# Article

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# Dielectric Control of Motor Fuel Compounding Plants

Nabil Abdel Sater<sup>1</sup>, Andrey Grigorov<sup>1</sup>, Gelena Neustroieva<sup>1</sup>, Oleksandr Bondarenko<sup>2</sup>, Vasyl Matukhno<sup>2</sup>, Sergii Vavreniuk<sup>2</sup>

<sup>1</sup> National Technical University «Kharkov Polytechnic Institute», 61002, 2 Kirpichova str., Kharkov, Ukraine <sup>2</sup> National University of Civil Defense of Ukraine, 61023, 94 Chernyshevskaya str., Kharkov, Ukraine

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#### Abstract

The article proposes the use of operational dielectric control system to increase the efficiency of operation of automatic compounding of motor fuels. These plants are used at oil refining enterprises in Ukraine and are an integral part of the technological chain of the commercial fuels production. It is established that all the main components and additives used for the production of commercial gasoline brands A-92, A-95 and A-98 have higher values ( $\varepsilon_{mix}$ ) than straight run base gasoline. And this, in turn, can be used for operational control of the gasoline compounding process. This control can be carried out on the basis of the information received from capacitive sensors which are located in pipelines of the main material streams. Moreover, the control is carried out on the content of components (X, %) or on the value of the octane number (ON, point) of the mixture on the basis of experimentally obtained dependences  $\varepsilon_{mix} = f(X)$  and ON =  $f(\varepsilon_{mix})$ .

**Keywords**: Motor fuel; Compounding; Operational control; Additives; Dielectric constant; Octane number; Properties; Gasoline.

#### 1. Introduction

Modern motor fuels are produced in accordance with the developed recipes and are a mixture of components obtained by technologies of primary refining of crude oil, secondary thermo-catalytic processes and additives for various functional purposes. They usually contain more than 200 individual hydrocarbons of different structure, the content of which as well as their interaction with each other determines the properties of the fuel. In the conditions of oil refineries motor fuels are obtained using an automatic compounding system, which is the final technological stage of production of commercial fuels. During compounding, the ratio of the components (their concentration) to be mixed and the quality of the commercial fuel obtained as a result of this operation are strictly controlled.

Today, control over the technological process of compounding is carried out on the basis of the results of laboratory analysis in the central factory laboratory of samples taken at the appropriate installation places. Note that laboratory control includes the definition of a number (from 3 to 5) of standardized quality indicators that characterize the properties of the fuel obtained. On our opinion, such a system is outdated and does not correspond to modern trends in improving the efficiency of operation of installations and technological equipment of Ukrainian refining enterprises.

#### 2. Research objective

Radical changes in this area should be made in the direction of development and implementation of modern systems of operative control of the technological process, which is based on obtaining the operational information. Such information is obtained when using in the system of quality the analyzers (sensors), which are located on the main material flows <sup>[1]</sup>. And on the basis of this information, adjustments to the process of fuel compounding are immediately made. When using the system of automatic control of motor fuels compounding the positive factors include:

1) improving the quality of marketable products;

- 2) preventing the release of substandard product;
- 3) reducing the costs for laboratory analytical control;
- 4) the ability to correct quickly the dosed input of components and additives.

It is clear that these factors can be met only by using for product quality assessment a single integrated indicator, instead of a set of standardized indicators (density, resistance to detonation, saturated vapor pressure, fractional composition, etc.), which adequately characterizes the composition and properties of motor fuel. Given the electrical properties of petroleum products (they are not polar dielectrics), such indicator is their relative dielectric constant ( $\epsilon$ ). Today, based on this indicator, a number of methods allowing to quickly obtain the information about the composition, properties of oil and petroleum products have been developed <sup>[2-4]</sup>.

#### 3. Results and discussions

For the production of standard motor fuel, for example, motor gasoline brands A-92, A-95 and A-98, usually equal proportions of straight-run gasoline fraction (b.p.-180 °C), catalytic cracking and reforming gasoline, supplemented with additives - substances that have high resistance to detonation are used <sup>[5]</sup>.

The effect of any component of gasoline on the value ( $\epsilon_{mix}$ ) of the resulting mixture in general can be represented as

#### $\varepsilon_{\text{mix.}} = \varepsilon_{\text{b.f.}} + \varepsilon_{\text{comp.}}$

(1)

where  $\varepsilon_{b.f.}$  – the value of the base (straight-run) gasoline fraction;  $\varepsilon_{comp.}$  – component value.

In this case, from the sources <sup>[6-7]</sup> it is known that there is a certain ratio between the main groups of hydrocarbons (paraffins, olefins, naphthenes, cycloolefins, aromatic hydrocarbons) which all petroleum products consist of

 $\varepsilon_{\rm p.} < \varepsilon_{\rm ol.} < \varepsilon_{\rm nap.} < \varepsilon_{\rm c.ol.} < \varepsilon_{\rm ar.}$ 

(2)

The chemical composition of straight-run gasolines is mainly represented by paraffinic and naphthenic hydrocarbons. Also, they contain a small, up to 5 % part of aromatic compounds <sup>[8]</sup>. In view of this, their value ( $\epsilon_{mix}$ ) does not exceed 1.9 units.

Catalytic cracking gasolines are represented by 52-55 % olefinic hydrocarbons, 39-43 % by paraffinic hydrocarbons and 5-6 % by aromatic compounds <sup>[9]</sup>. Due to this, their value  $(\epsilon_{mix})$  will be slightly higher than that of straight-run gasolines.

Catalytic reforming gasolines are 70-75 % represented by aromatic hydrocarbons, 25-30 % by a mixture of paraffinic and naphthenic hydrocarbons <sup>[10]</sup>. As a result, their value ( $\epsilon_{mix}$ ) will be higher than that of straight-run gasolines and catalytic cracking gasolines.

Then, when adding any of the components presented above to the straight-run gasoline fraction, there will be a certain increase of value ( $\epsilon_{mix}$ ), which is confirmed by the experimental data presented in Fig. 1.

At the maximum content of components in the mixture at the level of 35 % (see Fig. 1): for catalytic cracking gasoline  $\Delta \epsilon = 0.05$  units; for catalytic reforming gasoline  $\Delta \epsilon = 0.22$  units.

Today, the alcohols are increasingly used in gasoline compositions: methanol, ethanol, propane and iso-butane. This, in turn, helps to increase the environmental friendliness of gasoline - in the exhaust gases of the internal combustion engine there is a decrease in the amount of CO, NO<sub>x</sub> and solid C <sup>[11]</sup>. In addition, alcohols exhibit high antiknock properties. Usually the amount of alcohol in gasoline does not exceed 15 %, which increases the resistance to detonation by 6-8 points <sup>[11-12]</sup>.

The value ( $\epsilon$ ) of alcohols ranges from 17.7 to 33.1 units <sup>[6-7]</sup>, but when they are added to the straight-run gasoline fraction as well as in the case of catalytic cracking and reforming gasolines, the value ( $\epsilon_{mix}$ ) increases, that is shown on Fig. 2.





Figure 1. Dependence of  $(\epsilon_{mix})$  of the mixture on the content of component X: 1 - catalytic reforming gasoline; 2 - catalytic cracking gasoline

Figure 2. Dependence of  $(\epsilon_{mix})$  of the mixture on the alcohol content X: 1-methanol; 2 - ethanol; 3 - propanol; 4- iso-butanol

On value of ( $\epsilon$ ) the alcohols can be arranged in the following order: methanol > ethanol > propanol > iso-butanol. This is the dependence (see Fig. 2) that persists for mixtures of straight-run gasoline fraction with the studied alcohols. Thus, at the maximum alcohol content in the mixture at the level of 15 %: for methanol  $\Delta\epsilon$  = 2.46 units; for ethanol  $\Delta\epsilon$  = 2.33 units; for propanol  $\Delta\epsilon$  = 2.27 units; for iso-butanol  $\Delta\epsilon$  = 1.93 units.

To increase the detonation resistance of gasolines in addition to alcohols esters (dimethyl ether (DME), methyl tert-butyl ether (MTBE)) and N-methylaniline (MMA) are used.

Ethers, like alcohols, help to improve the environmental properties of gasolines (reducing CO emissions by up to 20 %), their starting properties and increase their resistance to detonation (OC = 98-103 points per motor method). The optimal concentration of esters in gasoline varies between 5-15 % <sup>[13-14]</sup>.

Also, in the composition of some gasolines such an additive as MMA can be found. Usually its optimal amount in gasoline is up to 2 %, which increases the resistance to detonation by 6 points. Although, according to <sup>[15]</sup>, it is forbidden to use it in Euro 5 gasoline.

Considering these additives in view of their influence on the value ( $\epsilon$ ) of the straight-run gasoline fraction, we note that DME and MTBE contain an oxygen atom (O), and MMA is a compound of cyclic structure with a nitrogen atom (N). This, in turn, contributes to a slightly larger (4.0-6.0 units) value ( $\epsilon$ ) than value of straight-run gasoline (up to 1.90 units). Therefore, the increase of the concentration (X, %) of these additives in the composition of the straight-run gasoline fraction will lead to an increase of the value ( $\epsilon_{mix}$ ) in comparison with the original straight-run gasoline fraction, which is illustrated in Fig. 3.

Applying the methods of mathematical statistics to the dependences presented in Fig. 1-3, we obtain regression equations that adequately ( $R^2=0.8234\div0.9982$ ) characterize the dependence ( $\epsilon_{mix}$ ) on X for the investigated components / additives (see Table 1).

Also of practical interest is the dependence of detonation resistance, expressed in points of octane number (ON) of commercial gasoline, on their ( $\epsilon_{mix}$ ). Based on the data presented in <sup>[3, 16]</sup>, this dependence has the character shown in Fig. 4. This dependence is used in the development of portable express commercial motor fuels quality analyzers <sup>[17-18]</sup>.

Component/Additive	Regression equation ( $p = 0,95$ )	R <sup>2</sup>
Catalytic cracking gasoline	$\varepsilon_{mix} = 0,0013 \times X + 1,8759$	0,8234
Catalytic reforming gasoline	$\epsilon_{mix} = 0,0066 \times X + 1,8796$	0,9819
Methanol	$\varepsilon_{mix} = -0,0031 \times X^2 + 0,2251 \times X + 1,7597$	0,9847
Ethanol	$\varepsilon_{mix} = -0,0024 \times X^2 + 0,2097 \times X + 1,7078$	0,9488
Propanol	$\varepsilon_{mix} = -0,0007 \times X^2 + 0,1675 \times X + 1,7684$	0,9786
Iso-butanol	$\varepsilon_{mix} = -0,0011 \times X^2 + 0,1569 \times X + 1,7668$	0,9852
ММА	$\varepsilon_{mix} = 0,0365 \times X + 1,8858$	0,9969
MTBE	$\varepsilon_{mix} = -0,0005 \times X^2 + 0,028 \times X + 1,8886$	0,9725
DME	$\varepsilon_{mix} = -0,0005 \times X^2 + 0,0187 \times X + 1,8848$	0,9982

Table 1. Regression equations for dependences obtained
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Figure 3. Dependence of  $(\epsilon_{mix})$  on the content of additives X:1 - MMA; 2 - MTBE; 3-DME

Figure 4. Dependence of octane number of commercial gasolines on their ( $\varepsilon_{mix}$ )

When using the above components / additives for the production of commercial gasoline the dependences shown in Fig. 1-4 can be used for compounding control. Then, the compounding scheme, which allows obtaining commercial gasoline brands A-92, A-95 and A-98, will have the form shown in Fig. 5.



Figure 5. Scheme of automatic compounding of motor gasoline

The gasoline compounding plant is located on a special production place, which has a fence around the perimeter and consists of three blocks (tanks block of components, tanks block of additives and compounding block), operator, software, power shield, control and measuring devices.

The tanks block of components and additives consists of tanks, filters, technological pipelines. The compounding block is represented by pump-dosing complexes, filters, pumps, control sensors and pipelines.

The functionality of the plant is the following: the main components of the tanks (T-1, T-2, T-3) pass through filters (F-1, F-2, F-3), where they are cleaned of mechanical impurities and

pump-dosing complex (PDC-1) feds them to the disperser (DIS-1). The dosing of the components is done automatically in strict accordance with the developed recipe by weight or volume. In (DIS-1) a compounding (mixing) of basic components occurs. Then the mixture is pumped by the pump (P-1) through the filter (F-4) and the capacitive sensor (S-1). Thermocompensation is used in the S-1 sensor to reduce the measurement error ( $\Delta X$ ). Finally, in the software the dependences presented on Fig.1 can be used for control of technological process.

The control of the parameter ( $\varepsilon_{mix}$ ) is carried out according to the equation  $\varepsilon_{mix} \approx \varepsilon_{st} \pm \Delta X$  (3)

where  $\varepsilon_{st}$  – standard value defined for a mixture of a certain composition.

If equation (3) is not fulfilled, the mixture is returned to (DIS-1) by means of valves along the technological pipeline. In this case, there is an automatic correction of the dosed input to the mixture, which is in (DIS-1) of the main components.

If equation (3) is satisfied, the mixture then enters the disperser (DIS-2), where mixing of basic components with additives supplied by (PDC-2) from tanks (T-4, T-5, T-6) through filters (F-5, F-6, F-7). Then the mixture of components with additives - commercial gasoline is pumped by the pump (P-2) through the filter (F-8) and the sensor of the finished product (S-2). The operation principle of the sensor (S-2) is similar to the principle of the sensor (S-1). But, at this stage, the software monitors the process either by the content of components (see Fig. 2-3) or by the value of the ON (see Fig. 4) of the obtained gasoline. For that the equation similar to equation (3) but obtained for commercial gasoline is used.

If equation (3) is not fulfilled, motor gasoline is returned to (DIS-2) by means of valves along the process pipeline. In this case, there is an automatic correction of the dosed input to the mixture, which is in (DIS-2) additives.

In case equation (3) is fulfilled, commercial gasoline is removed from the installation to the finished product warehouse or to the overflow overpass.

# 4. Conclusions

The scheme of the plant of motor gasolines automatic compounding in which the operative control of technological process is carried out taking into account electric properties of gasolines (dielmetric method) is offered.

The compounding scheme consists of three blocks (tanks block of components, tanks block of additives and compounding block) and the operational control system is represented by software and a number of sensors located in pipelines on key material flows (at the output of dispersers).

Operational control of compounding is offered to be carried out on the content of components (X, %) or on the size of octane number (ON, point) of mixture on the basis of experimentally received dependences  $\varepsilon_{\text{mix}} = f(X)$  and  $ON = f(\varepsilon_{\text{mix}})$ .

The presented scheme of automatic compounding and the system of its operative control can be used at production of jet and diesel fuels of various types, boiler fuel with the improved operational properties, and also lubricating oils of various functional purpose.

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To whom correspondence should be addressed: Dr. Andrey Grigorov, National Technical University «Kharkov Polytechnic Institute», 61002, 2 Kirpichova str., Kharkov, Ukraine; <u>e-mail: grigorovandrey@ukr.net></u>

# Article

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A Model for Predicting Choke Performance in Niger Delta

Christopher Iwowari, Joseph Amieibibama, Joseph Atubokiki Ajienka

Department of Petroleum and Gas Engineering, University of Port Harcourt, Nigeria

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#### Abstract

Chokes are very vital components of the producing system used to regulate flow and mitigate undesirable release of hydrocarbons to the environment from downhole. Basically, there are two types of chokes, the surface and subsurface chokes. The surface chokes are designed to operate at critical flow where pressure perturbations downstream are not transmitted upstream whereas the subsurface chokes are designed to operate at subcritical conditions where pressure perturbations downstream are transmitted upstream. This work focused on the development of a model for predicting surface choke performance using an Excel Solver- non-linear gradient reduction technique. With this solver, new sets of constants were developed for an existing choke performance model that was selected amongst several other correlations. Comparison of this model with the selected model and other existing models shows that the model from this study is most accurate in predicting choke performance. Statistical parameters were also used to ascertain the goodness of fit and agreement with existing models and the results show very good agreement.

Keywords: Multiphase flow; Choke performance; Critical flow; Subcritical; Correlations.

#### 1. Introduction

Multiphase flow in pipes refers the concurrent flow of liquid, gas and solid together in a pipe. Multi-phase flow is often characterized by liquids and gases occurring simultaneously, howbeit, with some solids in most cases. Virtually all flow phenomena in the petroleum production operations are multi-phase since no fluid is so clean, it does contain at least microscopic particles <sup>[1]</sup>. Multiphase flow occurs in many industries. Common examples where it occurs are in the nuclear industry, chemical industries, and naval engineering. It is mostly used to investigate phase interactions and hydrocarbon accounting <sup>[2]</sup>. Multiphase flow through restrictions in the oil and gas industry is of high importance to the production engineer and the two basic restrictions encountered are flow through well head chokes and subsurface safety valves (SSSV) <sup>[3]</sup>.

Well head chokes are used to maintain the well allowable production rate, control sand production by maintaining sufficient back pressure to protect surface equipment <sup>[4]</sup>. Chokes are also used for protection from other problems such as slugging, restricting flow rate and water coning <sup>[5]</sup>. Likewise subsurface safety valves (SSSV) are also important as they automatically shut in a well in cases of wellhead and/or surface equipment failure <sup>[6]</sup>.

The use of these restrictions is governed by subcritical and critical flow theory. Well head chokes operate on the principle of critical flow while subsurface valves operate on the principle of subcritical flow. Flow is critical when the velocity of the fluid through the restriction is sonic or attains a Mach number of 1(one). The surface chokes are usually designed and selected so that fluctuations in the pressure downstream of the choke have no effect or transmitted upstream while the subsurface chokes are designed to ensure that downstream perturbations are transmitted upstream <sup>[7]</sup>. Flow is subcritical when the fluid velocity is subsonic and fluid velocity is less than local velocity of sound <sup>[4]</sup>.

## 2. Previous works

Analytical and empirical approaches have been proposed to predict the multiphase flow behavior through wellhead chokes. Although, very few works have been done in subcritical flow, Gilbert <sup>[8]</sup> pioneered the empirical work that predicted critical flow through wellhead chokes, and is given as:

$$q_l = \frac{P_1^E d^D}{A R^B \gamma_0^C}$$

(1)

where: $q_l$  = gross liquid rate; BPD,  $p_1$  = wellhead pressure; psia, R = gas liquid rate (GLR) in Mcf/bbl.; d = Choke size in 64<sup>th</sup> of an inch;  $\gamma_o$  = oil gravity.; A, B, C, D and E are constants.

This correlation is the most popular choke performance correlation upon which many researchers had benchmarked the development of new correlations. However, one major limitation of the Gilbert's empirical correlations is that it assumes a fixed point for critical flow.

Many researchers have come up with various modified versions of the Gilbert's correlations. These modifications are centered on the values of the constants (A, B, C, D and E) in Equation 1 and the inclusion of additional variables while maintaining the general form of Equation 1. Examples of similar but modified versions of the Gilbert's correlation are those proposed by Different values of the constants for Equation 1 have been proposed by different authors such as Baxendell <sup>[9]</sup>; Ros <sup>[10]</sup>; Achong <sup>[11]</sup>; Pilehvari <sup>[12]</sup>, Al-Attar <sup>[13]</sup>, Owolabi *et al.* <sup>[14]</sup>, Khorzoughi *et al.* <sup>[15]</sup> and Okon *et al.* <sup>[16]</sup> respectively. Al-Attar <sup>[13]</sup> modified the Gilbert equation by including the API gravity of the crude but recommended the use of more field data to test the correlation. Al-Towailib and Al-Marhoun <sup>[17]</sup> introduced the effect of gas-oil mixture density into an empirical correlation that describes fluid flowing through chokes.

Ajienka and Ikoku<sup>[2]</sup> also developed a generalized analytical model. Their model is applicable to both critical and subcritical multiphase flow and can be used for continuous liquid phase and continuous gas phase and is given as:

$$q_{tp} = F_b \beta R_{mp} F_{mp}(X)$$

(2)

where:  $q_{tp}$  is the two-phase flow rate;  $F_b$  is the base factor;  $\beta$  is the multiphase beta factor;  $R_{mp}$  is the multiphase specific volume factor; and  $F_{mp}(X)$  is the dimensionless pressure factor.

Choubineh *et al.* <sup>[18]</sup> developed a critical multiphase flow model through chokes. The authors introduced additional useful variables, instead of the normal three variables used by other authors to improve the accuracy in determining critical flow rate. Their analytical model is based on data from south Iran and consists of choke size, wellhead pressure, gas specific gravity, temperature, gas liquid ratio and critical flow rate. Artificial neural network (ANN) with a teaching-learning based optimization (ANN-TLBO) algorithm was used in the development of their model and a non-linear regression approach was adopted to ascertain the values of their empirical constants and is given as:

$$Q_{I} = \frac{A \times P_{wh} \times D_{64}^{B} \times \gamma_{o}^{C} \times \gamma_{g}^{D} \times (\frac{T}{T_{sc}})^{E}}{\frac{1}{T_{sc}}}$$

(3)

where,  $P_{wh}$  is the wellhead pressure (psia);  $D_{64}$  is the choke size (1/64);  $\gamma_g$  is the gas specific gravity;  $\gamma_o$  is the oil specific gravity; T is the temperature;  $T_{sc}$  is the standard temperature (77°F); *GLR* is the gas-liquid ratio (SCF/STB), and  $Q_L$  is the liquid critical flow rate (STBD).

Joshua *et al.* <sup>[19]</sup> also developed a new correlation which is empirically based from the works of Choubineh *et al.* <sup>[18]</sup> method. Using 283 surface production data sets from 7 oilfields in Niger Delta, Joshua *et al.* <sup>[11]</sup> developed and validated two new models for predicting choke performance. The models are a function of the following variables: choke size, flowing wellhead pressure, oil specific gravity, gas specific gravity, surface temperature and gas liquid ratio. A non-linear multivariate regression was used to derive regression constants and exponents for the two models as shown in Equations 4 and 5.

$$Q_{L} = \frac{P_{wh}D_{64}^{C}}{AGLR^{B}}$$
(4)  
where  $A = 49.2531$ ;  $B = 0.4768$  and  $C = 2.230$  and  

$$Q_{L} = \frac{P_{wh}^{G}D_{64}^{C}(\gamma_{O})^{D}(\gamma_{g})^{E}(\frac{T}{T_{SC}})^{F}}{AGLR^{B}}$$
(5)  
where  $A = 0.7486$ ;  $B = 0.3066$ ;  $C = 1.6082$ ;  $D = 4.3376$ ;  $E = 0.0021$ ;  $F = 0.8999$ ;  $G = 0.6260$ .

# 3. Materials and methods

In this work, 2654 raw production data from 100 wells from 7 onshore assets in Niger Delta were obtained to develop a model for choke performance. The raw production data was cleaned to remove outliers using moving averaging technique to obtain a data structure suitable for regression analysis. These data is composed of total liquid rate, net oil flow rate, water flow rate, total gas flow rate, gas-oil ratio (GOR), gas-liquid ratio (GLR), tubing head pressure (psia), well flow line temperature (°F), wellhead choke size ( $d_{64}$ ), basic sediments and water (BSW), API gravity (@ 60 °F), and the flow line pressure.

A performance evaluation was first conducted on some correlations using these data from Niger Delta with statistical parameters to select the most accurate model that fits these data set. Thereafter, the selected correlation was used as a base for the development of the new correlation. An Excel Solver, a non-linear gradient reduction technique was used to determine the constants for the new correlation. The new correlation was then used to predict the pressure and compared with the base correlation and other correlations by means using error analysis.

#### 4. Results and discussions

As can be seen in Table 1, the most accurate correlation that best predicted the data set is Okon *et al.* <sup>[17]</sup> with an APE of 39% and standard deviation of 0.9229 compared to other correlations. Hence, it was selected as the base model for the development of the new correlation in this work. The Okon *et al.* <sup>[16]</sup> correlation is of the form:

$$P_{wh}^{E} = \frac{1/A(q_{liq})(GLR)^{B}(API)^{C}}{d_{64}^{D}(1-BSW/_{100})^{F} (T/_{TSC})^{G}}$$

(5)

where A, B, C, D, E, F and G are constants.

Table 1. Statistical evaluation of some selected correlations

Error	Okon <i>et al.</i> [16]	Gilbert <sup>[8]</sup>	Baxendell [9]	Khourzoughi et al. [15]
APE (%)	39	95	94	99
Standard deviation	0.9229	1.4581	1.4456	1.5992

Using the non-linear gradient reduction technique, a new sets of constants were developed for Equation 5 as shown in Table 2. Figure 1 shows a cross-plot of predicted and measured wellhead pressure values for Okon *et al.* <sup>[16]</sup> and this study. Data points on the diagonal implies exact and accurate predictions of the measured pressure, data points above the diagonal implies over-prediction while data points below the diagonal under-prediction of the measured data. As can be seen in Figure 1, model from this study better predicted the wellhead pressures than predictions from Okon *et al.'s* model. To further justify this, a statistical error analysis was performed on both models as shown in Table 3. As can be seen, all the three statistical measures, APE, standard deviation and R<sup>2</sup> values indicates that predictions from the model developed in this study performs better than that of Okon *et al.* Hence, the constants in the original Equation 5 can be replaced with the new constants developed in this study for evaluating choke performance of wells in Niger Delta.

Table 2. Values of constants for Equation 5 for Okon *et al.* <sup>[16]</sup> and the model developed in this work

Correlations	А	В	С	D	E	F	G
Okon <i>et al.</i> [16]	0.0509	0.6749	0	1.8133	1.321	0.2235	0.000029
This work	1.249	0.517	1.503	-0.421	3.863	0.54	-0.37709

Table 3. Statistical evaluation of base correlations and that in this study

Error	Okon et al. [16]	This study
APE (%)	39	16
Standard deviation	0.9229	0.3684
R <sup>2</sup>	0.6221	0.822



Figure 1. Cross plot of measured and predicted pressures measured for Okon *et al*. <sup>[16]</sup> and this study. The graph plots values of pressure from field data against values gotten from calculation using model from this study



Figure 2. A graph showing a cross plot of measured and predicted pressures for model in this study and other existing correlations



Figure 3. The graph compares results gotten from the model in this study as and field data

More so, the model from this study was compared with other choke performance models as shown in Figure 2. Again, from the cross-plot, the model from this study outperformed the others. In order further validate the accuracy this model, this model was tested with field data on a cross-plot as shown in Figure 3. Obviously, from the cross-plot in Figure 3, there is a strong correlation in the prediction of measured pressure values as the data set cluster around the diagonal.

# 5. Conclusions

A new model for evaluating choke performance for Niger Delta is developed. This model was developed using Okon et al model as a base model, hence identical except that the values of the constants are different and generated from an Excel Solver- non-linear gradient reduction technique. The model was tested with other models and shown to be more accurate in predicting choke performance compared other models. Also, to ensure it validity, it was tested with a field data and it showed good agreement with field data and other models. This models when is so far tested with Niger Delta and thus should be calibrated when to be deployed in regions outside the Niger Delta.

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To whom correspondence should be addressed: Dr. Joseph Amieibibama, Department of Petroleum and Gas Engineering, University of Port Harcourt, Nigeria; <u>e-mail: amieibibama.joseph@uniport.edu.ng</u>

# Article

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Research and Development of A Mathematical Model of a Polymer-Based Viscous Non Newtonian Fluid for Oil and Gas Wells Drilling

D. Rymchuk<sup>1</sup>, N. Shevchenko<sup>1</sup>, A. Tulska<sup>1\*</sup>, V. Ponomarenko<sup>1</sup>, O. Shudryk<sup>2</sup>

<sup>1</sup> National Technical University "Kharkiv Polytechnic Institute", Kyrpychova str.2, Kharkiv, Ukraine <sup>2</sup> Private limited company "Region", Kharkiv, Ukraine

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#### Abstract

Analysis of the use of biopolymers in drilling and well stimulation for oil and gas production has been performed. Normal Newtonian fluid model, taken used as a sample, is considered a nonlinear viscous fluid. Models that are most common in oil and gas industry are considered. Laboratory studies of rheology of an aqueous solution with various concentrations of xanthan gum have been carried out. An analysis of experimental and numerical hydrodynamic studies in flexible pipes of coiled tubing units has been performed considering the rheology of drilling fluid. For the numerical simulation of the hydrodynamics of a non-Newtonian fluid authors apply an open integrable platform OpenFOAM.

Keywords: Non newtonian fluid; Polymers; Drilling; Flushing.

# 1. Introduction

Nowadays, use of polymer additives for technical fluids in the oil and gas industry is of interest. Polymer additives can affect the rheological properties of aqueous and hydrocarbon systems and form gels of various densities, viscosities and rheology. Such compositions are widely used due to their high cleaning, transporting and retention properties. Long chain polymers are also capable of reducing hydraulic resistance in turbulent flows. Also, polymer so-lutions prevent the penetration of liquid filtrate into the pores of the treated rocks <sup>[1-2]</sup>.

Consider the use of biopolymers in drilling and well stimulation. The basis of technology for drilling oil and gas wells is cleaning the bottom of the wells with flushing fluid <sup>[3-4]</sup>.

Development of drilling technology and increase in the share of hard-to-recover reserves are followed by the evolution of drilling fluids so as modern multicomponent and multifunctional systems appear <sup>[5-6]</sup>. The study <sup>[7]</sup> presents an overview of polymer drilling fluids and a classification of polymer reagents used in drilling fluids.

Therefore, the most suitable materials for drilling fluids and well stimulation include a weighting agent such as clay powder, starch, polyanionic cellulose of low and high viscosity, xanthan gum (biopolymer), etc.

Nowadays biopolymer and xanthan-based flushing solutions are widely known: Flo-Pro systems from M-I Drilling Fluids Co and ANCO-2000, from ANCOR Drilling Fluids. Xanthan gum is a polysaccharide that dissolves in water, it is produced by microorganisms Xanthomonas campestris <sup>[8]</sup>.

Xanthan gum is used to regulate the rheological properties of water-based drilling fluids (both fresh and highly saline). It makes it possible to increase the viscosity even of low-concentrated solution, the gum provides its retaining and carrying characteristics. The recommended concentration of xanthan gum is 0.6 to 8 g/L.

#### 2. Information review

In the classical formulation rheological properties of a viscous incompressible Newtonian fluid is described by a linear equation <sup>[9]</sup>:

# $\tau = 2\mu S$ ,

where  $\mu$  – dynamic (molecular) viscosity of a Newtonian fluid;  $S \equiv S_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_i} + \frac{\partial u_j}{\partial x_i} \right)$  – strain

rate tensor;  $u_i, u_j$  – Cartesian components of vector velocity.

Some fluids are characterized by nonlinear viscosity, plasticity, elasticity – they are called non-Newtonian fluids. These non-Newtonian fluids are thoroughly described by rheological theory. For one class of fluids that have similar properties, a generalized rheological model is presented. The simplified model of the studied fluid is described by the generalized Newtonian model, in which the fluid is considered a nonlinear viscous fluid with the effective viscosity [10-14]. Effective molecular viscosity depends on the rate of deformation of the fluid  $\dot{\gamma} = \sqrt{2S.S.}$ 

The generalized Newtonian model for a viscoplastic non-Newtonian fluid has the form:  $\tau = \mu_{eff}(\dot{\gamma}) \cdot \dot{\gamma}$ (2)

Models of fluid with a power-law rheological law, Bingman fluid and Herschel-Bulkley fluid are the most common in the oil and gas industry [5-6,10-17]. Effective viscosity is described by the following equations:

$\mu_{eff}(\dot{\gamma}) = k_{\nu}\dot{\gamma}^{n-1} - \text{ rate model};$	(3)
$\mu_{eff}(\dot{\gamma}) = \frac{k_v \dot{\gamma} + \tau_0}{\dot{\gamma}}$ – Bingman model;	(4)
$\mu_{eff}(\dot{\gamma}) = rac{k_{v}\dot{\gamma}^{n}+ au_{0}}{\dot{\gamma}}$ – Herschel-Bulkley model.	(5)

where  $k_{\nu}$  is a measure of the fluid consistency; n is the indicator of the non-Newtonian environment;  $\tau_0$  is the ultimate shear stress (yield point).

Generalized information on the rheology and hydrodynamics of non-Newtonian fluids is presented in <sup>[14]</sup>. Laboratory studies of the rheological parameters of a drilling fluid with polymer additives for drilling permafrost are presented in <sup>[15]</sup>. The rheology of solutions is described by the Power-law and Bingham models.

A description of the new type of flushing fluid FLO-PRO is given in [16]. The results of laboratory and industrial studies of a biopolymer system for drilling inclined and horizontal boreholes of deep oil and gas wells are presented. An experience of long-time well drilling has shown that clay-free biopolymer solutions are effective for technical, economic and safety reasons.

In <sup>[17]</sup>, experimental and computational studies of the effect of polymer additives on the resistance to the movement of liquids in pipelines are presented. It is noted that a small concentration of polymer additives to water and oil products impart new rheological properties to the fluid due to which the hydraulic resistance in turbulent flow is sharply reduced (Toms effect).

The results of numerical studies of the non-Newtonian models of a viscous fluid flow in a cylindric pipe, annular gaps, channels of hydraulic machines are presented in [13-14,18-19].

The aim of the work is to increase the efficiency of well drilling and intensify oil production by using biopolymer flushing solutions based on xanthan gum. The main tasks are considered: study of the rheological properties of an aqueous solution at various concentrations of xanthan gum; analysis - experimental and numerical hydrodynamic studies in flexible pipes of coiled tubing installations, taking into account the rheology of the drilling fluid.

# 3. Experimental studies of the rheological properties of the solution at various concentrations of xanthan gum

The current work is devoted to study of rheology of a water-based fluid mixed with xanthan gum biopolymer with a concentration of 1.2-12 g/L with a step of 1.2 g/L. The structure of the test solution is determined using a rotary viscometer. The initial data and results are presented in Table. 1. The magnitude of the shear stress is determined for eight values of the rotor speed of the viscometer - viscometer rotor speed (VRS, min<sup>-1</sup>) and ten values of the concentration of xanthan gum (CXG, g/l). We have assumed that the rate of deformation of the fluid (shear rate) is identical to the rotor speed of the viscometer.

	Concentration of xanthan gum, g/L									
VRS, min <sup>-1</sup>	1.2	2.4	3.6	4.8	6	7.2	8.4	9.6	10.8	12
3	1	3	6	9	12	16	18	25	30	38
6	2	3	6	9	14	18	20	28	34	40
30	3	4	8	13	17	23	28	34	42	51
60	5	6	10	15	20	26	34	38	46	57
100	5	7	11	17	22	29	35	42	50	62
200	7	9	16	21	27	36	40	49	60	74
300	8	11	17	25	32	40	48	56	70	83
600	10	16	22	32	38	50	58	68	84	98

Table 1. Results of laboratory study of shear stress magnitude  $\tau_{0}$ , Pa

The rheological relation between the shear stress and the rotor speed of the viscometer are shown in Fig. 1. Results are shown for five concentrations of xanthan gum.



Fig. 1. Rheological characteristics of the studied process fluid

Processing laboratory studies of rheology. Determination of effective viscosity. The rheological lines on the Fig. 1 indicate that the studied liquids are described by the rate model  $\tau(\gamma) = k_{\nu} \cdot \gamma^n$ . The coefficients of the rheological model  $k_{\nu}$  and n were defined by approximating laboratory data. The results are shown on the Fig. 2.





At n=1 and  $k_v = 1$  mPa·s, the concentration of the gum is zero, i.e. the studied fluid is water. Using formula (3) the values of effective viscosity of the liquid can be defined. The effect of effective viscosity on the shear rate for five values of the concentration of xanthan gum in an aqueous solution are shown on the Fig. 3.



Fig. 3. The effect of rotor rotation speed of effective viscosity at various concentration of xanthan gum in water

Results of the study show that increase of the rotor speed of the viscometer (shear rate) is followed by decrease of viscosity for all concentrations of xanthan gum. These solutions are pseudoplastic fluids. The effect of effective viscosity on the concentration of xanthan gum shows that at low shear rates, the viscosity of the solution has a minimum at a concentration of 1.2 g/L. Further increase of xanthan gum concentration is followed by increase the viscosity of the solution. The results of rheology studies are used to determine the flow regime in pipes and predict the hydraulic resistance coefficients.

#### 4. Experimental studies of flushing solution supply through a flexible coiled tubing pipe

In this work there has been studied pumping of a xanthan gum (XG) in water solution through a flexible pipe of a coiled tubing installation. The studies have been carried out at 11 operating modes of the pumping unit (Q-fluid flow through the pipe in L/min). Also, as a result of the experiment, the type of pumped liquid changed. The concentration of xanthan gum biopolymer varied from 1.2 g/L to 12 g/L. As a result of the tests, the required fluid injection pressure was determined. Coiled tubing pipe outlet pressure is equal to atmospheric pressure. Therefore, it is assumed that the injection pressure is equal to the hydraulic losses along the length of a pipe. Table 3 shows the test results: the effect of concentrations of xanthan gum, the flow of the pumping unit and the pressure loss in atmospheres when pumping liquid through a 38.1 mm pipe with a wall thickness of 3.2 mm and a pipe length of 5620 m.

Fluid flow Q,				Concenti	ration of x	kanthan g	jum , g/L			
L/min	1.2	2.4	3.6	4.8	6	7.2	8.4	9.6	10.8	12
50	29.4	40.1	63	90.5	114.4	147.7	174.3	205.4	250.1	300
80	33.1	40.9	75.4	106.7	132.4	169.8	200.4	231.8	281.8	336.3
100	39.2	45.7	61.6	116.2	142.8	182.6	215.3	246.8	299.8	356.6
120	46.7	52.1	68.4	92.6	152.6	194.4	229.1	260.7	316.3	375.3
150	60.4	64.3	79.9	104.5	122.9	154.7	248.4	280	339.2	401
180	77.1	79.4	93.3	118.2	135.5	167.6	196.3	217.7	360.5	424.9
200	89.9	91	103.3	128.3	145	177.2	206.5	226.2	273.4	440.1
230	111.6	110.9	119.9	145.1	160.7	193.1	223.2	240.5	288.2	337.1
250	127.8	125.9	132.8	157.3	172.3	204.8	235.2	251.1	299.1	347.7
280	154.7	150.7	155.4	177.4	191.3	223.9	255.1	268.6	317.1	365.2
300	174.4	169	171.9	191.9	205	237.7	269.4	281.3	330.1	377.9

Tab. 3. Pressure loss in a flexible pipe at different modes of pumping liquid with different concentration of xanthan gum, atm

# 5. Results and discussion

According to hydraulics laws <sup>[10]</sup>, the pressure loss in a cylindrical pipe for a viscous fluid is determined by the equation:

$$\Delta P = \lambda \left(\frac{L}{D}\right) \frac{\rho V^2}{2}$$

(8)

where  $\lambda$  is the hydraulic resistance coefficient; D is the hydraulic diameter of the pipe; L is the pipe; V is the average velocity of the fluid in the pipe;  $\rho$  is the density of the liquid.

While pipe length L and diameter D are constants, fluid velocity V may vary and depends on the operating mode of the pump unit, i.e. V = f(Q). Pressure losses are known from the experiment and depend on the operating mode of the pump and the type of pumped liquid - $\Delta P = f(Q, k_v, n)$ . Based on the results of experimental studies, we calculate the coefficient of hydraulic losses  $\lambda$  according to the formula:

$$\lambda(\mathbf{Q}, \mathbf{k}_{v}, n) = \frac{2D \cdot \Delta P(\mathbf{Q}, \mathbf{k}_{v}, n)}{L \rho \cdot V(Q)^{2}}$$

(9)

Effect of the resistance coefficient on the flow rate of the pumped liquid is shown on the Fig. 4.



Fig. 4. Effect of the resistance coefficient in the pipe on the operating mode of the pumping unit at the concentration of xanthan gum in water: 1 - 1.2 g/L; 2 - 2.4 g/L; 3 - 3.6 g/L; 4 - 4.8 g/L; 5 - 6 g/L; 6 - 8.4 g/L.

According to the hydraulic law, the drag coefficient is represented as a function of the Reynolds criteria. Non-Newtonian fluids are described by generalized Reynolds criteria. For a fluid described by the rate rheological model, the Metznel-Reed Reynolds criteria expression is normally used <sup>[10, 12]</sup>:

$$Re^* = \frac{D^n V^{2-n} \rho}{\frac{k_v}{8} \left(\frac{6n+2}{n}\right)^n},$$

where  $k_{\nu}$  – measure of a fluid consistency; n – non-Newtonian area indicator.

Results of recalculation of the experimental data for determination of  $\lambda = f(Re^*)$  are presented on the Fig.5. Analysis of the experimental data of the drag coefficient in a coiled tubing pipe for a non-Newtonian fluid shows that increase of the biopolymer concentration in water provides the laminar mode of the fluid flow. The pressure loss coefficient decreases as the concentration raises up.

For mathematical modeling (MM) of a turbulent flow of an incompressible viscous fluid, a system of continuity and Navier-Stokes equations is used <sup>[9, 13]</sup>:

$$\begin{cases} \frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_j} (\rho u_j) = 0; \\ \frac{\partial}{\partial t} (\rho u_i) + \frac{\partial}{\partial x_j} (\rho u_i u_j) + \frac{\partial}{\partial x_j} (\rho u'_i u'_j) = -\frac{\partial p}{\partial x_i} + \frac{\partial}{\partial x_j} \left[ \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \right] + f_i, \end{cases}$$
(6)

where  $i, j = 1 \dots 3$  - summation over the same indices;  $x_1, x_2, x_3$  - coordinate axes;  $u_1, u_2, u_3$  - time-averaged velocities;  $u'_1, u'_2, u'_3$  - pulsation components of velocities; fi - expresses the effect of mass forces.



Fig. 5. Changing of the resistance coefficient  $\lambda = f(Re^*)$  at various concentration of xanthan gum in water: 1 - 1,2 g/L; 2 - 2,4 g/L; 3 - 3,6 g/L; 4 - 4,8 g/L; 5 - 6 g/L; 6 - 8,4 g/L.

To close the system of equations (6), the two-parameter Menter SST model is used <sup>[20]</sup>. Menter's model is written by a superposition of the  $k-\varepsilon$  and  $k-\omega$  models, based on the fact that the  $k-\varepsilon$  type models describe the properties of free landslide flow better and the  $k-\omega$  model they have an advantage when modeling near-wall flow. A smooth transition from the  $k-\omega$  model in the near-wall region to the  $k-\varepsilon$  model for zones far from solid walls is provided by introducing weight empirical functions <sup>[20, 13]</sup>.

To confirm the reliability of coefficient calculation of the rate rheological model of a non-Newtonian fluid, a numerical simulation of a three-dimensional viscous flow was carried out using OpenFOAM software system <sup>[21]</sup>. The system is based on a set of libraries that provide tools for solving systems of partial differential equations in both space and time. The working language of the code is C ++. In terms of this language, most of the mathematical differential and tensor operators in the program code (before translation into the executable file) equations can be presented in a readable form, and the discretization and solution method for each operator can be selected by the user during the calculation.

The calculated area is taken to be the volume of liquid filling the inner space of a flexible pipe 1 m long and 38 mm in diameter.

A mesh of computational domain was created by the block method, a computational mesh with hexahedral cells and a wall layer was constructed to ensure the value  $y + \leq 2$  [21-22].

The velocity vector in m/s was set as a boundary condition in the inlet section. The calculation was carried out for the operating range of the pump Q: 50 - 300 L/min. At the exit from the computational domain, the static pressure was set equal to  $P/\rho = 101.325 \text{ m}^2/\text{s}^2$ .

A comparison of the experimental and calculated pressure losses along the length of the pipe is shown in Fig. 6. The numerical results are recalculated for the operating conditions of the coiled tubing. Consistence of the experimental data is observed for aqueous solutions with xanthan gum concentration up to 6 g/L.



Fig. 6. Comparison of the experimental and calculation pressure loss along the pipe length. Concentration of xanthan gum:1 – 2,4 g/L; 2 – 6g/L; 3 – 8,4 g/L; 4 – 12 g/L.  $\Delta$  – calculation points received by the *OpenFOAM* 

#### 6. Conclusions

Research of polymer additives in process fluids have shown that solution based on biopolymer additives is considered a non-Newtonian fluid. A structure of viscous non-Newtonian rate model fluid based on xanthan gum in a coiled tubing coiled tubing has been studied.

The basic equations of the mathematical model of stationary motion of a generalized Newtonian fluid do not differ from the classical model of a Newtonian fluid. In turbulent flows, the molecular viscosity of a non-Newtonian medium depends on the fluctuating shear rate. Therefore, when describing a turbulent flow, it is necessary to operate with the averaged value of the effective molecular viscosity.

Modeling a three-dimensional viscous flow of a non-Newtonian viscous fluid shows an adequate similarity between the experimental and calculated data, which indicates the correct choice of the mathematical model, the turbulence model and the correctness of determining the coefficients of the power-law model of the fluid and.

The research results make it possible to predict the value of the drag coefficient and pressure loss when pumping a viscous non-Newtonian fluid through a flexible coiled tubing pipe.

To improve the accuracy of design work, it is necessary to carry out preliminary laboratory studies of the rheological parameters of the test fluid (drilling fluids, water-oil mixtures, solutions with polymers.

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To whom correspondence should be addressed: Dr. A. Tulska, National Technical University "Kharkiv Polytechnic Institute", Kyrpychova str.2, Kharkiv, Ukraine; <u>e-mail: fuel.khpi@gmail.com</u>

# Article

Laboratory-Based Correlations to Estimate Geomechanical Properties for Carbonate Tight Reservoir

Nagham Jasim Al-Ameri

Petroleum Engineering Department, University of Baghdad, Iraq

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#### Abstract

Rock mechanical properties are critical parameters for many development techniques related to tight reservoirs, such as hydraulic fracturing design and detecting failure criteria in wellbore instability assessment. When direct measurements of mechanical properties are not available, it is helpful to find sufficient correlations to estimate these parameters. This study summarized experimentally derived correlations for estimating the shear velocity, Young's modulus, Poisson's ratio, and compressive strength. Also, a useful correlation is introduced to convert dynamic elastic properties from log data to static elastic properties. Most of the derived equations in this paper show good fitting to measured data, while some equations show scatters in correlating the data due to the presence of Calcite, Quartz, and clay in some core samples. Brittleness index (BRI) indicates ductile behavior of the core samples is also studied for the interested reservoir. The results of BRI show that the samplers range from moderate to high brittleness, and the difference in BRI comes from the presence of some minerals, as explained using the X-ray diffraction test (XRD). The proposed correlations are compared to other correlations from literature for validation, and the results of the comparison show good matching that explains the accuracy of the proposed equations.

**Keywords**: Static elastic properties; Dynamic elastic properties; Brittleness index; X-ray diffraction test (XRD); Compressive strength; Experimentally-derived correlations.

#### 1. Introduction

In recent studies, geomechanical properties are important parameters to eliminate many problems in the oil industry. For instance, estimating geomechanical properties during drilling can reduce wellbore instability by providing the failure criteria around the wellbore [1-2]. Wellbore instability problems can be reduced significantly when the study is related to wellbore stability changes during reservoir depletion [3-5]. Other problems related to well drillings, such as sand production, can be eliminated by accurately determining geomechanical properties [6-7]. Relating geomechanical properties to hydraulic fracturing propagation is another essential study proposed by many authors such as Huang *et al.* [8] and Al-Dossary *et al.*, [9].

In unconventional reservoirs, the importance of implicating geomechanical studying in production and development stages becomes essential. Developing a tight reservoir required specific strategies due to the ultralow permeability of these reservoirs.

Laboratory-based determination of geomechanical properties is the accurate method to use, even though it is expensive and time-consuming. When the core samples for the depth of interest are not available for testing, it is recommended to derive empirical correlations from the available core samples measurements. The obtained correlations will be beneficial to find geomechanical properties such as Poisson's ratio (v), Young's Modulus (E), and compressive strength (CS). Another essential step after geomechanical properties measurements is to derive an equation that relates the dynamic and static properties [10-12]. Developing such direct correlations between dynamic and static geomechanical properties can effectively estimate mechanical properties from log data in the depth of lacking measurements.

This study uses geomechanical and physical properties measurements of 12 core samples to find significant experimentally derived correlations for a carbonate reservoir. These derived equations are essential in drilling, hydraulic fracturing, and enhancing recovery for the tight reservoir under study. Also, the obtained relations were compared to other relations reported in the literature to explain the validity of the derived correlations.

#### 2. Samples properties

In the current study, 12 core samples are collected from three wells (wells B, C, and V5X) passing through the studied carbonate reservoir and the samples cover the main sections of the reservoir. The reservoir consists of two main layers: A and B layers. A layer consists of mud lime without hydrocarbon shows. While the B layer is a hydrocarbon show, its estimated initial oil in place (IOIP) represents 25% of the total IOIP in the studied field. The layer B consists of three main layers, which are B1, B2, and B3.

The primary lithology of B1 layer is bioturbated wackestone, while the B2 layer comprises primarily bioturbated packstone and pelagic foram chondrites, which are either dolomitized or pyrite cemented. The B3 layer has developed shale containing smectitic, pyrite, oolitic packstone, skeletal intraclasts packstone; echinoderms, pebbles, oolitic, and dark grain (pyritic), and chondrite can also be found in the layer B3.

The depths, geomechanical, and petrophysical properties measurements of the core samples are listed in Table 1. The sample's physical properties include compressional velocity (Vp), shear velocity (Vs), bulk density ( $\rho_b$ ), and porosity ( $\varphi$ ). The geomechanical properties of the samples are also listed in table-1, including Poisson's ratio ( $\upsilon$ ) and Young's modulus (E) measurements in both static and dynamic methods (ultrasonic test). All the testing methods and the proposed formula for the determination of mechanical properties for the core samples are mentioned by <sup>[13]</sup>. The laboratory results data listed in Table 1 will be used to find general correlations between physical and geomechanical properties for studied reservoir.

	Sample no./layer	Depth m	ρ₀ gm/cc	Poros- ity%	E <sub>s</sub> , Gpa	Us	Ud	E₀, Gpa	C.S., Gpa	Vs, m/s	V <sub>P</sub> m/s
Well B	1/S-B1	2673	2.67	18.73	19.98	0.330	0.25	15.568	73.509	3149	3643.6
	2/S-B2	2694	2.67	23.4	17.23	0.257	0.25	19.099	57.871	3148	3644
	3/S-B3	2711	2.68	24.58	5.406	0.157	0.25	14.129	52.402	3143	3636
Well C	4/S-B1	2735	2.66	15	12.92	0.257	0.25	12.818	70.575	2936	3400
	5/S-B2	2762	2.66	20.7	12.28	0.242	0.25	15.382	58.224	2935.7	3402
	6/S-B3	2796	2.71	19.8	5.056	0.255	0.25	11.699	42.345	2930	3392
	7/S-B1	2686.33	2.66	19.21	18.04	0.27	0.29	19.00	173.94	3202	3722
Well	8/S-B1	2680.68	2.66	19.21	12.83	0.28	0.29	13.00	163.96	3205	3720
V5X	9/S-B2	2701.71	2.65	25.5	13.88	0.28	0.29	13.00	141.39	3193	3614
	10/S-B2	2722.66	2.66	23.7	15.98	0.30	0.32	18.00	188.43	3190	3611
	11/S-B3	2725.33	2.68	25.78	7.91	0.22	0.25	10.00	130.19	3190	3620
	12/S-B3	2728.07	2.68	25.6	5.35	0.14	0.25	10.00	118.86	3190	3622

Table 1. Core samples properties and depth

# 3. Empirical equations

Two general methods are known to evaluate rock mechanical properties. One method involves estimating stress-strain behavior by applying different load range on a rock sample. This method allows for measuring the static elastic properties of the formation. The other method involves measuring both compressional and shear wave propagation velocities used to estimate dynamic rock elastic properties using fundamental relations. In obtaining these relations, samples of high clay content will be neglected to reduce the misfit in data plotting and obtain a systematic equation to represent the required relations. This section will illustrate the main outlines for adopting these two methods for the reservoir under study.

## 3.1. Relation between compressional and shear velocity

Figure 1 shows the linear relation between the compressional and shear velocity of the carbonate 12 samples. The obtained equation's high correlation factor (0.9936) reveals the strong relationship between the two velocities. The obtained equation is given below:  $V_s = 0.8222V_p + 142.84$  (1)

Due to the absence of shear velocity in conventional logging data, the obtained experimentally derived equation can be beneficial to approximate the shear velocity of the reservoir at any depth.



Figure 1. Relation between compressional and shear velocity

# **3.2. Estimating dynamic mechanical properties and conversion from dynamic to static elastic properties**

Young's modulus and Poisson's ratio indicate rock deformation. When the estimated Young's modulus is high and Poisson's ratio is low, then this formation is more rigid. The dynamic Young's modulus and Poisson's ratio are usually derived from well logs and are calculated using the measured  $V_p$  and  $V_s$  as follows:

$$v_d = (V_P^2 - 2V_S^2)/2(V_P^2 - V_S^2)$$
  

$$E_d = [\rho_b V_S^2(3V_P^2 - 4V_S^2)/(V_P^2 - V_S^2)]$$

(2) (3)

(4)

(5)

where  $E_d$ =dynamic Young's modulus;  $\rho_b$  =density (g/cm<sup>3</sup>);  $V_s$ =Shear wave velocity (m/s);  $V_p$ =compressional wave velocity (m/s), and u= Poisson's ratio.

Figure 2 explain the linear relation between static and dynamic Poisson's ratio and the relation between static and dynamic Young's modulus. The following equations are obtained from linear elastic properties relation of the studied reservoir:

 $E_s = 1.3244E_d - 7.4154$  $v_s = 1.2v_d - 0.076$ 





The static elastic modulus is the most important in rock mechanics because it reveals rock deformation under high applied stress. Sometimes it is not easy to conduct a laboratory measurement to obtain static properties. Therefore, these correlations are essential to convert dynamic Young modulus and Poison's ratio to approximated static Young modulus and Poison's ratio.

#### 3.3. Obtaining compressive strength correlations

The compressive strength (CS) of a rock can be measured directly in the laboratory. This parameter indicates rock hardness. Experimental measurements of CS represent the maximum load that the sample was subjected to before fracturing. Compressive strength can be calculated using velocity or mechanical properties. Compressional velocity, shear velocity, and Young modulus listed in the Table 1 have been used in this paper to estimate compressive strength. Figure 3-a demonstrates a positive relation between compressional velocity and compressive strength.



Figure 3. Relation between compressional, shear velocity, and compressive strength

In contrast, Figure 3-b illustrates the positive relation between shear velocity and compressive strength. The obtained relationships have an excellent fitting and are very useful to find CS for different ranges of both shear and compressional velocities values. The obtained correlations are given in the following equations:

 $CS = 1.3172V_p - 4734.9$ 

 $CS = 2.5124V_s - 7849.9$ 

(6) (7)

The relationship between compressive strength CS and Young's modulus for the studied reservoir is illustrated in Figure 4. The data are scattered, and it is challenging to obtain a relation between compressive strength CS and Young's modulus.



Figure 4. Relation between dynamic Young's modulus and compressive strength

In practice, using Young's modulus to estimate compressive strength is not straightforward. Therefore, the obtained relation will be either overestimate or underestimate the CS, while using compressional or shear velocity to estimate CS is a direct relation. The scatter shown in Figure 4 for the estimated CS can be attributed to the diagnosis process of some core samples such as calcite, quartz, and cement. These minerals are found in the studied core samples due to the X-ray diffraction (XRD) test conducted for the 12 core samples. According to the XRD test of the core samples, the calcite percentage is higher, and it ranges from 76.2% to 90% of the total sample mineral.

On the other hand, the total clay is the second higher mineral percentage after calcite, and it ranges from 7.3% to 15.2% of the total mineral content according to the XRD test. In contrast, Quartz represents the third higher third mineral content in the interested samples within a range of 4.24% to 5.5%. The presence of these minerals within a higher percentage in the core samples effects on core's porosity and the accuracy of log data of these cores. This mineral effect on samples' porosity will affect the calculated dynamic Young's modulus. Therefore, the resulting relation between CS and Ed has not fit the data.

The same mineral content range and types (calcite, quartz, and cement) in the core samples cause a wide scattering when finding a relation between compressive strength CS and cores porosity. Figure 5 shows such high data scattering and suggests that porosity alone is not a good indicator for compressive strength estimation. The difficulties in finding a beneficial relationship between rock compressive strength and porosity are also mentioned by <sup>[14]</sup>. The author indicates that the most empirical equations relating to rock physical properties and strength do not fit the measured data.



Figure 5. Relation between core porosity and compressive strength

#### 3.4. Rock brittleness calculation

Tight reservoir development needs specific strategies. One of the typical strategies to increase tight reservoirs recovery is hydraulic fracturing. In hydraulic fracturing operations, rock brittleness is a vital parameter to reflect the ability of the rock to create fractures. If the rock is brittle, then long fractures are expected. The ductile behavior of the rock indicates the difficulty of inducing hydraulic fracturing.

In general, brittleness can be affected by the lithology of the rock, especially quartz and clay content <sup>[15]</sup>. Another critical parameter affecting the degree of rock brittleness is the rock elastic properties. Rock's high brittleness indicates high Young's modulus and low Poisson's ratio. In this study brittleness index for the studied core samples are calculated based on both Young's modulus and Poisson's ratio <sup>[16]</sup> as follows:

$BRI\_E = \frac{E - E_{min}}{E_{max} - E_{min}} * 100$	(8)
$BRI_{v} = \frac{m_{v-v_{max}}}{v_{min} - v_{max}} * 100$	(9)
$BRI_avg = 0.5*(BR_E+BR_v)$	(10
where DDI D, builtlances index based on Verrage medul	

where: BRI\_E: brittleness index based on Young's modulus; E: measured value of Young's modulus, GPa; E\_min: minimum value of Young's modulus, GPa; E\_max: maximum value of Young's modulus, GPa; BRI\_v: brittleness index based on Poisson's ratio; v: measured value

of Poisson's ratio;  $\upsilon$  \_max: maximum value of Poisson's ratio;  $\upsilon$  \_min: minimum value of Poisson's ratio; BRI\_avg: brittleness index based on Young's modulus and Poisson's ratio.

For the studied core samples, the calculated brittleness index (BRI) and its relation to elastic properties are illustrated in Figure 6-a. A perfect relation is obtained to find BRI in terms of Poisson's ratio using the following equation: BRI = -526.32 v + 173.68 (11)

On the other hand, no relation is obtained to estimate rock brittleness in terms of Youn's modulus, as shown in Figure 6-b. Grieser *et al.*, <sup>[17]</sup> stated that if the brittleness index is higher than 40, the rock could be considered brittle, and if the value is higher than 60, then the rock is very brittle.



Figure 6. Relation between core brittleness and elastic properties

# 4. Case study and validation of the new correlation



Figure 7. Calculated and measured mechanical properties

The well log data from well B were used for estimating mechanical properties for the reservoir using the proposed correlations in the current study. Figure 7 shows the compressional transit time log from well B. The shear transit time is not available for this well, so equation 1 is used to obtain shear velocity and convert it to shear transit time. The results of calculating shear transit time are drawn in Figure 7. The measured shear and compressional time data from Table 1 are also drawn in Figure 7 as dot points to explain the validation of the used equation. Then these two velocities are used to calculate both dynamic Young's modulus and dynamic Poisson's ratio using equations 2 and 3, respectively. The new derived correlations (equations 4 and 5) are then used to convert the dynamic elastic properties to static ones, and the results are shown in Figure 7. The measured Young modulus and Poisson's ratio are also drawn in Figure 7 to illustrate the accuracy of equations 4 and 5 in dynamic to static conversion of elastic properties.

The compressive strength is calculated for well B using the new correlation derived in

the current study. Using shear and compressional velocity, equations 6 and 7 are used to obtain the compressive strength. The calculated compressive strength and the measured CS from Table 1 are drawn in Figure 7 to illustrate the high accuracy of the new correlations.

Another critical step is to check the accuracy of the proposed correlation by comparing the new correlation (reservoir correlation) results to the available correlations in the literature. Equation 1 for estimating the correlation between Vs and Vp for S-reservoir is compared with Castagna correlation and Ameen *et al.* correlation since these correlations are given for the same lithology of carbonate S-reservoir. Castagna correlation is given in equation 12 <sup>[18]</sup>, and Ameen *et al.*, <sup>[19]</sup> correlation are given in equation 13.

$$V_S = 0.4403 V_P + 0.576$$

 $V_S = 0.52V_P + 0.25251$ 

(12) (13)

The results of the comparison of the three equations are shown in Figure 8 to indicate an accepted result of the proposed new correlation. The difference in using current correlations (equation 1) and other correlations (equation 12 and 13) for the same carbonate reservoir is expected. This difference is due to the difference in formations porosity between the low porosity of tight reservoir samples and the sample's higher porosity of other correlations.



Figure 8. Comparison of Vs and Vp correlations

The new correlation of estimating rock compressive strength introduced in equation 6 using compressional velocity Vp is compared with the correlation proposed by Militzer and Stoll <sup>[20]</sup> as given in equation 14. The results of Equations 6 and 14 are shown in Figure 9.  $CS = 2.45V_p^{1.92}$  (14)



Figure 9. comparison of compressive strength correlations

# 5. Conclusions

Measured mechanical properties were used in the current study to obtain important correlations, which can be used for other carbonate reservoir. The proposed equation works well in fitting the measured data to estimate reservoir mechanical properties. Scatter in some relations in obtaining good fitting to measured data comes from certain minerals in core samples. It is essential to mention that the obtained correlations are proposed for low porosity- low permeability samples, so caution should be taken when comparing the new correlations to other correlations.

The brittleness index is calculated based on Young's modulus and Poisson's ratio. The calculated brittleness index is an essential tool to estimate rock's ability to fail without compressive strength measurements. A perfect relation is obtained to find BRI in terms of Poisson's ratio while no relation is obtained to relate BRI to Young modulus.

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To whom correspondence should be addressed: Nagham Jasim Al-Ameri, Petroleum Engineering Department, University of Baghdad, Iraq; <u>e-mail: naghamjasim@coeng.uobaghdad.edu.iq</u>

# Article

# **Open Access**

Synergistic Effect Between Some Copolymers and Organic Solvents to Improve the Flow Properties of Waxy Crude Oil

#### Taisir Taha Khidr

Department of Petroleum Applications, Egyptian Petroleum Research Institute (EPRI), Cairo, Egypt

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#### Abstract

Wax deposition is a common problem in oil pipelines and production systems. The modification of the wax crystal habit is useful in the transportation and processing of crude oil at low temperatures. Various pour point depressant (PPD) additives can help with this modification via a variety of mechanisms. Hexadecyl acrylate and NAFOL 1822B acrylate are synthesised in this work by reacting a hexadecyl alcohol/NAFOL 1822B alcohol blend with acrylic acid, and two copolymers are synthesised by copolymer hexadecyl acrylate with vinyl acetate and another copolymer NAFOL 1822B acrylate with methyl methacylate. They characterized by IR spectroscopy and GPC was used to calculate their molecular weights. These copolymers were tested with various solvents for their efficacy as a crude oil pour point depressant and flow improver. The solvents utilised in this study were xylene, n-hexane, kerosene, and triethanolamine (TEA) for the wax dispersing wax operation. The effect of different percentages of these solvents with copolymer additions in the crude oil sample on the crude oil flow characteristics was assessed. All of the solvents tested enhanced crude oil flow properties. Blends of kerosene and triethanolamine (TEA) with the copolymer NAFOL 1822B acrylate (NA) -methyl methacrylate (MMA) were more effective than the other solvents as a crude oil pour point depressant. *Keywords: Copolymers; Solvents; Additives; Pour point depressant; Viscosity; Crude oil.* 

#### 1. Introduction

Transportation of crude oil is considered as a difficult and highly technical operation. To deal with the high viscosity, which is one of the main challenges in pipeline transportation <sup>[1-2]</sup>, more efficient and commercial methods are required It is well known that wax crude oils have a high viscosity and a high pour point, making production, transportation, and refining more difficult, especially at low temperatures <sup>[3-4]</sup>. As a result, different crude oil flow improvers have been used to improve heavy oil <sup>[5-6]</sup>. Dilution with lighter crudes or alcohols, as well as heating, are some of the common methods, but it depends on the feedstock of the lighter crudes, and the cost of using alcohols is very high. In general, these additives can be classified as crude oil flow improvers as long as they improve the flowability of crude oil. Pour point depressant and flow improver are the two most common types of crude flow improver. Pour point depressant and viscosity reduction are the two most prevalent types of crude flow improvers [<sup>7-8]</sup>.

Polymers with a wax-like paraffinic part and a charged component, such as polyalkyl acrylates and methacrylates, ethylene vinyl acetate copolymers, copolymers and homo of alpha olefins, alkyl fumarate-vinyl acetate copolymers, and alkyl esters of styrene-maleic anhydride copolymers <sup>[9-11]</sup>, are commonly used chemical additives. According to <sup>[12]</sup>, the process of oil refinement contains negative factors such as large molecular weight, long molecular chain, and high thermo stability of these polymers.

In this case, crude oil is small molecular additives are required to act as a viscosity reduction. The structural features of gelled waxy crude oils have recently been studied in order to resume oil pipeline transit <sup>[13-14]</sup>. The first goal of this research was to use a copolymer ingredient to improve the flowability of waxy crude oil and lower its pour point. As a result, a hexaacrylate-

vinyl acetate copolymer and a NAFOL 1822B acrylate-methyl methacrylate copolymer (hexaacrylate-vinyl acetate copolymer and NAFOL 1822B acrylate-methyl methacrylate copolymer) were prepared. The second goal was to investigate the rheological behavior of two copolymers in different solvents, as well as the pour point depression of this molecule when mixed with waxy crude oil.

# 2. Experimental

# 2.1. Materials

Aldrich chemical provided hexadecyl alcohol, methyl methacrylate (MMA), acrylic acid, vinyl acetate, p-toluene sulphonic acid, and toluene as analytical reagents, which were used exactly as received. Benzoyl peroxide, the initiator, was re-crystallized from methanol. CONDEA chemical company provided a linear long-chain alcohol blend and (NAFOL 1822 B) with the typical analysis listed in Table 1. Qarun Petroleum Company submitted Egyptian waxy crude oil (CO). Table 2 lists their physicochemical properties. Gas chromatography analysis was used to determine the n-paraffin distribution of the isolated waxes in accordance with ASTM D 2887 standards.

Properties	NAFOL 1822B			
	С <sub>16 - ОН</sub>	0.1		
	С <sub>18 - ОН</sub>	5.0		
	С <sub>20 – ОН</sub>	16.5		
Alcohol composition (wt.%)	С <sub>22 – ОН</sub>	77.6		
	С24 - он	0.8		
Average carbon number (calculated)	C <sub>av</sub> = 21.5~22			
Density (g/cm <sup>3</sup> ) approx.	at 80°C = 0.802			
Solidification point (°C) approx.	64			
Flash point (°C) approx.	204			
Ester No. (mg KOH/g)	0.14			
Acid No. (mg KOH/g)	0.05			
Iodine No. (mg I/100 mg)	0.31			
Water (Wt.%)	0.04	1		

Table 1. Typical analysis of linear long chain alcohol blends (NAFOL)

Table 2. Physical characteristics of waxy crude oil

Properties	Method	Crude oil
Specific gravity at 60/60°F	IP 160/87	0.8557
Kinematics viscosity at 35°C (cSt)	IP 71/80	15
Pour point (pp), °C	IP 15/67(80)	25
Sulfur content (wt%)	IP 266/87	0.21
Total paraffins content (wt%)	Urea adduct	20.75
n-paraffin (wt%)	GLC	20.05
Iso-paraffins (wt%)	GLC	0.70
Asphaltene content,wt.%	IP 143	00.728
Wax content, wt%	UOP 46/64	10.920

# 2.2. Synthesis of additive

# 2.2.1. Synthesis of monomers

Synthesis of n-alkyl acrylic acid (0.3 mol) was esterified with a series of n-alkanols with increasing number of carbon atoms (hexadecyl alcohol C16, and NAFOL 1822 B (Cav= 22) (0.2 mol) in a round bottomed flask equipped with a Dean and Stark trap to remove the reaction water azeotropically as it formed. The reaction was carried out at reflux temperature

in the presence of toluene (100 mL) as a solvent P-toluene sulfonic acid was used as a catalyst, and hydroquinone was used as a polymerization inhibitor. The two prepared monomers (hexadecyl acrylate and NAFOL 1822 B acrylate) were then purified by distilling off the solvent at the end of esterification at low pressure. The reaction mixture was then extracted with n-hexane, washed with distilled water to remove the catalyst and any unreacted acid until the washing became neutral, and the organic layer separated.

## 2.2.2. Preparation of copolymers

Two copolymers, P1 copolymer additive of hexadecyl acrylate (HA)-vinyl acetate (VA) and P2 copolymer additive of NAFOL 1822B acrylate (NA)-methyl methacrylate (MMA), were prepared by free radical polymerization in the presence of dibenzoylperoxide (0.5 wt percent) as an initiator, which was dissolved in toluene and added to the reaction mixture in equal metered portions every 0.5 hour during the course of the reaction. At the start of polymerization, the reaction mixture was nitrogen flushed for twenty minutes before being gradually heated to reflux temperature under a nitrogen blanket while adding the first portion of initiator and vigorously stirring. The reaction was allowed to run for 6 hours. They were purified by precipitating them in an excess of methanol and then filtering them. Purification was completed by dissolving in toluene and precipitating with methanol, followed by filtration and vacuum drying at 40°C.

# 2.2.3. Method mixed the solvent

The pour point test and kinematic viscosity were used to determine the efficacy of xylene, n-hexane, kerosene, triethanolamine (TEA), and their blends with copolymer additives as flow improvers for crude oils. Each improver, copolymer additives (P1,P2), xylene, n-hexane, kerosene, and triethanolamine (1 percent by volume) were added to the crude oil at room temperature (28°C). Each blend contains the following proportions: blend 1 is a mixture 0.5 mL of P1/P2, 0.5 ml of xylene and 0.5 mL of n-hexane, blend 2 contains 0.5 ml of P1/P2, 0.5 ml of p1/P2, 0.5 ml of kerosene), and blend 3 is a mixture of 0.5 ml of P1/P2, 0.5 ml of xylene, 0.5 mL of n-hexane and 0.5 mL of triethanolamine. Each blend was added to the crude oil at room temperature (28°C).

#### 2.3. Characterization of copolymers

In the technology laboratory, the structures of the prepared mono-esters hexadecylacrylate /NAFOL1822B acrylate and copolymers hexadecylacrylate with vinylacetate/NAFOL1822B acrylate with methylmethacrylate, abbreviated as P1 and P2, were confirmed using infrared (IR) spectroscopic analysis. The KBr technique was used to measure the infrared spectra with a model Genesis series (USA) infrared spectra-photometer. Table 3 depicts the characterization of the prepared copolymers.

Sample	Designated	Mol. wt.	PDI
Copolymer hexadecyl acrylate (HA)-vinyl acetate (VA)	P1	35,841	1,164
Copolymer NAFOL 1822B acrylate (NA) - methyl meth- acrylate (MMA)	P2	24,588	1,3784

Table 3. Molecular weights of the prepared copolymers

The molecular weight was determined in the technology laboratory using Gel permeation chromatography (GPC), which included a Waters 515 HPLC pump, a Waters temperature control module, and 2410 refractive index detector from Water Company. And further supplement using viscosity (Brookfield viscometer, USA).

#### 2.4.Evaluation method

# 2.4.1. Pour point measurement (ASTM D 97-96)

Effective the effectiveness of xylene, n-hexane, kerosene, triethanolamine (TEA), and their blends with the copolymer P1/P2 as flow improvers for crude oils was determined using the pour point test shown in Table 4, Figure 1. Each improver, xylene, n-hexane, kerosene, and

triethanolamine (1 percent by volume), was added to the crude oil along with the copolymers P1 and P2. . Blend 1 contains (0.5 mL of xylene, 0.5 mL of n-hexane, and 0.5 mL P1/P2), Blend 2 contains (0.5 mL of triethanolamine and 0.5 mL of kerosene), and Blend 3 contains (0.5 mL of xylene, 0.5 mL of n-hexane, 0.5 mL of triethanolamine, and 0.5 mL of P1/P2). At a low temperature, each blend was added to the crude oil.

Table 4. Effect of combined crude oil, copolymer additives and different solvents on pour point of crude oil

Additives (%)	Pour point (°C)	Pour point depres- sant (°C)
Crude oil	25	0
Crude oil+1%P1	20	5
Crude oil+1% P1+1% xylene	21	4
Crude oil+1% P1+1%n- hexane	20	5
Crude oil+1% P1+1% kerosene	15	10
Crude oil+1% P1+1% TEA	16	9
Crude oil+1% P1+1% xylene +1% n- hexane	18	7
Crude oil+1% P1+1% TEA +1% kerosene	14	11
Crude oil+1%P2	15	10
Crude oil+1% P2+1% xylene	19	6
Crude oil+1% P2+1%n- hexane	18	7
Crude oil+1% P2+1% kerosene	12	13
Crude oil+1% P2+1% TEA	13	12
Crude oil+1% P2+1% xylene +1% n- hexane	15	10
Crude oil+1% P2+1% TEA +1% kerosene	10	15



Figure1. Effect of combined crude oil, different additives and different solvents on pour point of crude oil

# 2.4.2. Determination of viscosity

The viscosity was calculated according to ASTM D 445-06. The kinematic viscosity of crude oil P1/P2 and the blend with different solvent was determined at 25°C and 35°C was used to containing the copolymers study the effect of different solvent, type of copolymer, and crude oil on kinematic viscosity. All of the experimental data is presented in Table 5 and Figures 2

and 3. The procedures were repeated for crude oil samples copolymer (P1,P2) without any solvent and treated copolymer additives (P1,P2) with 0.5 mL of xylene,0.5 mL n- hexane,0.5mL kerosene, and 0.5 mL triethanolamine (TEA), and then with the blends blends1 (0.5 mL of xylene and 0.5 mL of n-hexane), blends 2 (0.5 mL of kerosene and 0.5 mL of triethanolamine) and blends 3( 0.5 mL of xylene, 0.5 mL of n-hexane and 0.5 mL of triethanolamine). Each blend was added to the crude oil with the all the synthesized additive (P1 and P2).

Table 5. Effect of combined crude oil, copolymer additives and different solvents on viscosity of crude oil at different temperatures

Additives (0/)	Viscosity,cSt	
Additives (%)	At 25°C	At 35°C
Crude oil	40	15
Crude oil+1%P1	22	14
Crude oil+1% P1+1% xylene	20	14
Crude oil+1% P1+1%n- hexane	19	13
Crude oil+1% P1+1% kerosene	17	12
Crude oil+1% P1+1% TEA	15	11
Crude oil+1% P1+1% xylene +1% n- hexane	20	13
Crude oil+1% P1+1% TEA +1% Kerosene	12	10
Crude oil+1% P1+1% xylene +1% n- hexane+1% TEA	22	16
Crude oil+1% P2	20	14
Crude oil+1% P2+1% xylene	18	13
Crude oil+1% P2+1%n- hexane	17	12
Crude oil+1% P2+1% kerosene	16	12
Crude oil+1% P2+1% TEA	13	10
Crude oil+1% P2+1% xylene +1% n- hexane	15	12
Crude oil+1% P2+1% TEA +1% Kerosene	10	7
Crude oil+1% P1+1% xylene +1% n- hexane+1% TEA	18	12



Figure 2. Effect of temperature on viscosity of crude oil with P1 additive and different solvents





# 3. Results and discussion

# 3.1.Analytical methods

# 3.1.1. Molecular weight analysis

Table 3 shows the experimental values of average molecular weights (Mw) and polydispersity index data (PDI) of the prepared copolymers (P1 and P2). This result also indicates that polymer P2 is less polydispersed than polymer P1, implying greater uniformity. The values show that as the alkyl chain length of the alcohol used in esterification increases, the Mw values decrease while the PDI value increases.

#### 3.1.2. Spectroscopic analysis

#### - Ester monomers

Infrared spectral analysis was used to investigate the chemical structure, which revealed similar patterns for the two esters. The OH stretching band of the mono carboxylic group absorbed a small broad band at 3443 cm<sup>-1</sup> as a result of the partial esterification reaction in a representative IR pattern for NAFOL 822B acrylate monomer. The stretching vibrations of the CH<sub>3</sub> and CH<sub>2</sub> groups were strongly absorbed at 2913–2845 cm<sup>-1</sup>, the ester C=O appeared as a strong absorption band at 1707 cm<sup>-1</sup>, and the presence of ester functionality was further supported by the appearance of a C=O stretching vibration at 1154–1188 cm<sup>-1</sup>. The characteristic C=C stretching band was detected at 1636 cm<sup>-1</sup>, while the CH in plane deformation vibrations of acrylate ester monomers were detected at 1323 cm<sup>-1</sup>. A strong absorption of the bending 713 cm<sup>-1</sup>.

#### -Copolymers

In IR spectra was illustrated absence of peaks above 3000 cm<sup>-1</sup> indicates absence of C=C unsaturation which supports complete polymerization in all the cases. The characteristic C=C peak of alkyl acrylate vanished completely. Therefore, the P1 and P2 copolymers were synthesized successfully. Lack of peaks over 3000 cm<sup>-1</sup> in IR spectra demonstrates the absence of C=C unsaturation, which confirms full polymerization in all circumstances. As a result, the P1 and P2 copolymers were successfully produced.

# 3.2.Evaluation tests

# 3.2.1. Effect of copolymer additives and solvents on the pour point

The depression of the crude oil pour points is shown in Table 4 and Figure 1. P1 with crude oil lowered the pour point to 20°C, while P1 with Xylene lowered the pour point of crude oil to 21°C from 25°C. P1 with n-hexane decreased the pour point to 20°C, while P1 with kerosene

decreased it to 15°C, P1 with triethanolamine (TEA) reduced the pour point to 16°C, P1 with xylene and n-hexane blends reduced it to 18 °C and P1, triethanolamine, and kerosene blends reduced it to 14°C. While crude oil with P2 reduced the pour point to 15 °C, the additives may co-crystallize with the paraffin wax and modify their crystal structure. It was also discovered that there is a direct correlation between chain length and efficiency, with efficiency increasing as chain length increases, so the prepared additives were ranked as P1 <P2 <sup>[15]</sup>.

The prepared additives reduced the pour-point temperatures from 25°C to 15°C at the maximum pour-point reduction of 1000 ppm of P2. The pour point reduction <sup>[16]</sup> was calculated using the following equation: pour point reduction ( $\Delta PP$ ) = PP<sub>pure</sub>-PP<sub>add</sub>; where PP<sub>pure</sub> is the pour point of the crude oil without any additives (blank) and PP<sub>add</sub> is the pour point of the treated crude oil .

P2 with xylene reduced the pour point of the crude oil to 19°C, while P2 with n-hexane reduced it to 18°C. P2 with kerosene lowered the temperature to 12°C. P2 with triethanolamine (TEA) lowered the temperature of the oil to 13°C from 25°C. P2, xylene, and n-hexane blends reduced it to 15°C, while P2, triethanolamine, and kerosene blends reduced it to 10°C. The decrease in pour point is primarily caused by wax crystal modification. Pour point depressants molecules are adsorbed on different crystal faces P2 than P1, reducing the interlocking forces in P1 additive between two nuclei of wax molecules and deforming regular crystal growth <sup>[17]</sup>.

When compared to copolymers,P2 additive has a higher efficiency as PPD. It could be because P2 has a higher polarity than P1 additive. The highly polar polymers are highly adsorbed by the wax crystals present in crude oil, improving their efficiency as polymer PPD improvers <sup>[18]</sup>. When present in crude oil, the pour point depressant changes the wax crystal shapes from thin extensively interlocking plates to more compact crystals by co-crystallizing with the wax <sup>[19]</sup>. The triethanolamine (TEA) actively reduced the samples' pour points and wax deposition potentials after doping. The oxygen-containing group in triethanolamine inhibits wax growth and poisons it via an adsorptive surface poisoning mechanism <sup>[20]</sup>. The waxes then formed in the crude oil as small particles and were unable to form the net-like structure required for solidification and deposition.

#### 3.2.2. Effect of copolymer additives and solvents on the crude oil viscosity

Table 5, Figures 2 and 3 show how effective the added flow improvers are on the viscosity of the crude oil as well as their blends. At room temperature (25°C), the kinematic viscosity of the crude oil reduced from 40 cSt to 20 cSt and 18cSt for the crude oil containing P1 and xylene, P2 and xylene respectively, reduced to 19 cSt and17 cSt for crude oil containing P1 and n-hexane, P2 and n-hexane respectively, reduced to 17cSt and 16 cSt for crude oil containingP1 with kerosene, P2 with kerosene respectively and reduce to 15 cSt and 13 cSt for crude oil containing P1 and triethanolamine, P2 and triethanolamine, respectively. On adding copolymer additives with solvents blends, blend 1 (mixture of 0.5mL P1,0.5mL xylene and 0.5mL n-hexane/ mixture of 0.5 mL P2, 0.5mL xylene and 0.5 mL n-hexane), blend 2 (mixture of 0.5mL P1, 0.5mL triethanolamine and 0.5mL kerosene/mixture of 0.5mL P2,0.5mL riethanolamine and 0.5mL kerosene/mixture of 0.5mL P2,0.5mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine/mixture of 0.5mL P2, 0.5mL xylene,0.5 mL n-hexane and 0.5mL triethanolamine) to the pure crude oil, the viscosity of the crude oil at 25°C reduced. Blend 1 reduced the viscosity from 40 cSt to 20 cSt and 15 cSt, blend 2 reduced it to 12 cSt and 10 cSt blend 3 increased it from 40 cSt to 22cSt and18 cSt respectively.

As temperature increased, the viscosity of the crude oil reduced Table 5, Figures 2 and 3. This is in line with the reports by <sup>[21]</sup>, that crude oil response differently with the same additive at different temperature due to the changes in rheological properties of the crude oil. The Table 5 shows that the additive plays an important role in affecting the viscosity of the crude oil. The viscosity of the crude oil can be improved by adding requisite amount of flow improver, the appropriate type P1 or P2 and volume of the additive has to be added for effectiveness. From the results in Table 5 triethanolamine (TEA) is a very good wax deposition inhibitor with P2 additive than P1 additive. The reduction of viscosity on the addition P2 additive of these solvents is due to the dissolution of paraffin wax, which shows the effectiveness of these

additives, also effect of chain length where efficiency increase as chain length increase so P2 showed high viscosity reduction. Reduction of viscosity depends on the temperature type of additive and solvent.

# 3.3. Mechanism analysis

The low-temperature flow ability of crude oil is improved to better understand the P2 additive, and the possible working mechanism is depicted in Figure 4. Because n-alkanes have a high melting point in untreated crude, the wax crystals gradually precipitate, agglomerate, and interact with each other to form a three-dimensional network structure, losing fluidity at low temperatures. The long-chain alkyl of P2 additive inserts and co-crystallizes with n-alkanes in crude oil, providing an abundance of crystallization sites, thereby delaying the starting precipitation temperature, modifying the growth orientation of wax crystals, and preventing the formation of large crystals in P2 additive treated crude oil. As a result, the crystallization behavior and morphology of the crystal are modified, resulting in a better cold flow property illustrated in Figure 4.



Figure 4. Illustration for wax crystallization process of treated with P2 additive

#### 4. Conclusion

For crude oil, the copolymers can act as effective pour point depressants and flow improvers. Because of its longer alkyl chain, copolymer NAFOL 1822B-methyl methacrylate (P2) outperformed copolymer hexadecyl - vinyl acetate as a pour point depressant and viscosity reducer. The effects of different solvents xylene, n-hexane, kerosene, and triethanolamine on the pour point and viscosity of crude oil were investigated. All of the additives tested in this study were effective at lowering crude oil is pour point and viscosity of the crude oil. At different temperatures, triethanolamine and kerosene with P2 additive were more effective than triethanolamine and kerosene with P1 additive (25°C, 35°C).

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To whom correspondence should be addressed: professor Taisir Taha Khidr, Department of Petroleum Applications, Egyptian Petroleum Research Institute (EPRI), 1 Ahmed El-Zomor St., Nasr City, 11727, Cairo, Egypt; <u>e-mail: kkidrr@yahoo.com</u>

# Article

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Viscosity-Temperature Dependence Study of Crude Oil Emulsions from Some Oilfields in Niger Delta Region

B. J. Osiname 1, R. U. Duru 1, U. J. Chukwu 1\*, J. A. Ajienka 2

<sup>1</sup> Department of Pure and Industrial Chemistry, University of Port Harcourt, Port Harcourt, Nigeria <sup>2</sup> Department of Petroleum Engineering, University of Port Harcourt, Port Harcourt, Nigeria

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#### Abstract

Crude oil emulsion samples labelled W001, W002, W003, W004 W005 and W006 obtained directly from well-heads of two different oilfields in Niger Delta were characterized with respect to density, SG, BS&W, water cut and sulphur content. Viscosity-temperature dependence study was equally carried out at different temperatures ( $25^{\circ}$ C to  $90^{\circ}$ C) using OFITE 900 Digital Rotational Viscometer at 300 rpm and shear rate of  $510.69 \text{ s}^{-1}$ . Results obtained revealed that the crude samples were all sweet crude thus having low sulfur contents (0.120 to 0.316%wt) - a typical property of Niger Delta crude oil. Based on API categorization, one of the samples is heavy (W002 – 19.56), three are medium (W/003 – 23.36, W/004 – 23.36 & W/006 – 23.63) and the other two are Light (W001- 33.23 & W005 – 36.93). The BS&W values showed that sample W/003 had little amount of water molecules trapped in the crude (0.5%) while sample W/001 had high water molecules trappled in the crude sample (72%). The viscosity-temperature dependence study carried out at different temperatures ( $25^{\circ}$ C to  $90^{\circ}$ C) established that the viscosity of the emulsions changes with increasing temperature as molecules in the liquid phase gain energy to move faster. Also, the activation energy and the pre-exponential factor (entropic factor) of the samples were determined using the modeling of Arrhenius theory equation.

Keywords: Viscosity; Temperature; Crude oil emulsion; Oilfields; Niger Delta.

#### 1. Introduction

Crude oil is formed alongside gas and saline water known as formation water from the reservoir <sup>[1-2]</sup>. As the reservoir is being produced, it becomes depleted overtime with a corresponding decrease in the reservoir pressure, producing water alongside. The production of water alongside crude oil creates several problems during production <sup>[3]</sup>. The water may be produced either as free water which will later settle out of the mixture over-time or could form an emulsion (this could be a tight or loose emulsion).

These immiscible fluids are emulsified and the emulsification process is easily facilitated by the corresponding effect of shear and pressure drop at the wellhead, chokes, valves and any other restrictions along the flow path <sup>[4]</sup>. When this occurs, an emulsion is formed.

An emulsion is a combination of a two-phase system that is not homogenous when mixed and one is a disperse phase while the other is in the continuous phase <sup>[5]</sup>. In oil in water emulsion (O/W emulsion), the disperse phase is crude oil while the continuous phase is water. For a water in oil emulsion (W/O emulsion), the disperse phase is water while the crude oil is the continuous phase. According to report <sup>[6]</sup>, various types of emulsions are seen in different fields of technology such as the food industry, pharmaceuticals, and biological systems.

The formation of emulsion is possible during the transportation of crude oil. Hence, it is imperative to check the percentage of water (Base Sediment & Water – BS &W) in the crude oil before transportation. The conditions that support the formation and stability of emulsions include the presence of surfactants (surface acting agents), ionic compositions and pH of the water <sup>[4]</sup>. The stability of crude oil emulsions has become a major concern among researchers and this has led to formulating different methods of breaking it <sup>[4]</sup>. The formation of oilfield

emulsions which occurs from the reservoir has become complex and a herculean task to resolve in the oil and gas industries. If not properly and adequately resolved, emulsion formation could affect oilfield installations and processes in several ways which include: A high pressure drops in oilfield pipelines and flowlines, an upsurge in the cost of pumping and transportation of the emulsion (water/oil mixture) through the pumps and pipes. Also, damage of pipelines and other oilfield installations as a result of a di-phasic flow and the existence of chloride ions in the water phase. An increase in the rate of corrosion and scaling tendencies as a result of the presence of salts in the formation water thus the lowering of the API gravity of the crude oil <sup>[4, 7]</sup>.

Crude oils can be classified by categorizing them into four primary components which are: saturates (including waxes), aromatics, resins and asphaltenes. This classification of crude oils is commonly referred to as SARA fractionation and it is dependent on its polarity and solubility in a solvent <sup>[4]</sup>.

Viscosity is a property of fluid that indicates the resistance to flow. It is one of the physicochemical properties of fluids that is key to optimizing and designing industrial processes <sup>[8]</sup>. Viscosity is a property that is affected by temperature and pressure <sup>[9]</sup>. It thins with an increase in temperature and thickens as temperature decreases. The temperature-viscosity relationship of fluids plays an important role in many fields of technology [10]. The knowledge of viscosity is useful in fluids transport and petroleum industry [11-12]. In the manufacturing companies such as chemical, cosmetic, pharmaceutical etc., viscosity is critical for hydraulic calculations used for fluid transport and energy calculations <sup>[9]</sup>. Liquid viscosity needs to be measured because it affects several operational areas such as handling, transportation, injection, and combustion efficiency <sup>[9]</sup>.

According to published report <sup>[13]</sup>, it was established that temperature has a strong influence on the viscosity of fluid products with viscosity generally decreasing with increase in temperature. The relationship between the viscosity of a fluid and temperature has been established to be in the form of Arrhenius equation. The Arrhenius equation was first written in 1913 and the developed by Eyring in 1936 <sup>[8]</sup>. The Arrhenius equation is expressed as presented in equations 1 and 2. (1)

 $K = Ae^{-Ea/RT}$ 

 $\ln n = \frac{Ea}{RT} + \ln A$ 

where R the gas constant (8.31 JK<sup>-1</sup>mol<sup>-1</sup>); Ea, is the Arrhenius activation energy; A is the pre-exponential/Entropic factor. This can sometimes be called Arrhenius constant.

(2)

When the natural logarithm of viscosity  $(ln \eta)$  is plotted against the reciprocal of absolute temperature in Kelvin (1/T) for a liquid system, the graph is basically linear; while -Ea/R is the slope and *ln* A is the intercept.

#### 2. Materials and method

Six crude oil emulsion samples were obtained from different oilfields located within the Niger delta region of Rivers state, Nigeria. The samples were collected directly from the wellhead at their flow line pressures and labeled W/001, W/002, W/003, W/004, W/005 and W/006. Thereafter, they were shipped to the laboratory for preservation and further analysis.

All reagents used are of analytical grade and obtained from BDH. Basic characterizations were done on the samples. Density and specific gravity were done based on ASTM D1289 using hydrometer (HB-Instruments co.). Water cut (%vol/vol) by "Dean and Stark" distillation using ASTM D4007 method. The BS&W was done by ASTM D4006 with a centrifuge machine (Damon/IEC) and the sample pre-heated to  $60^{\circ}$ C in a thermostatic water bath (Mermmert). The sulphur content was determined according to ASTM D4294 using X-ray Fluorescence sulphur analyser (Horiba SLFA-2800). The sulphur analyser was previously calibrated before sample analysis with a standard oil of known sulphur content.

Rotational rheometry is a vital and powerful mechanism used for the measurement of complex shear rheology <sup>[3, 14]</sup>. In this study, the OFITE 900 Digital Rotational viscometer was used. The rheology test was carried out at 300 rpm and at various temperatures from 25°C to 90°C. The emulsion samples were later heated in a thermostatic bath (Memmert) and centrifuged in a centrifuge machine (Damon/IEC) at 600 rcf (relative centrifugal force) to separate the water in the emulsions from the crude oil and the recovered crude oil was characterized based on SG, density, API (ASTM D1289) and kinematic viscosity (using Stanhope-Seta KV-8 viscometer bath by ASTM D445).

## 3. Results and discussion

## 3.1. Characterization

Characterizations of the emulsion samples were measured, and the results listed in Table 1. The SG and density of the studied emulsion samples as shown in Table 1 obviously depict W/004 as the heaviest while W/005 is the lightest. W/005 and W/001 have very close densities. This observation is similar to the values obtained for W/003 and W/006. The sample W/004 has the highest BS&W value. This is indicative of the fact that the sample has a high percentage of water dispersed in a continuous phase of the crude oil than observed for other samples.

S/N	Sample ID	SG @ 60ºF	Density @ 15°C (g/cm <sup>3</sup> )	BS&W (%)	Water cut (%vol/vol)
1	W/001	0.8596	0.8570	7	72
2	W/002	0.9397	0.9388	20	30
3	W/003	0.9117	0.9108	0.5	6
4	W/004	0.9627	0.9618	40	50
5	W/005	0.8551	0.8542	17	60
6	W/006	0.9189	0.9161	22	59

Table 1. Values of specific gravity, density, BS&W and water cut of the crude oil emulsion samples

The water cut for the samples W/001, W/002, W/003, W/004, W/005 and W/006 are: 72%, 30%, 6%, 50%, 60% and 59% respectively. Sample W/001 has the highest water cut with 72%, while sample W/003 has the lowest with 6%. The implication of the high water cut is that the well where the sample was taken is producing more water than crude oil. Hence, for every 100 barrels of produced crude oil emulsion that flows out from the well-head, 72 barrels is water while 28 barrels is crude oil. It is understood that the viscosity of emulsion is affected by increasing the amount of water cut fractions <sup>[15]</sup>.

According to previous report <sup>[16]</sup>, any crude sample with sulfur content less than 0.5% is sweet crude. Samples with total sulfur content more than 0.5% are sour crude. Table 2 presents the weight % of sulfur content in the crude oil samples. All the samples measured have low sulfur content ranging from 0.12 to 0.31 which implies that all the samples fell under the classification of sweet crude. Sulfur and its compounds are very poisonous and dangerous to people, environment and assets (oilfield installations). It can lead to rust and could be the cause of corrosion of metals on oilfield installations. The presence of sulfur and its compounds have a very foul and offensive odour. Sulfur is a cause of worry to gas producers and refineries <sup>[16]</sup>. However, all six samples are sweet crude a characteristic of Niger Delta crude.

S/N	Sample ID	Sulphur content (wt. %)
1	W/001	0.23137
2	W/002	0.31625
3	W/003	0.21529
4	W/004	0.12000
5	W/005	0.1316
6	W/006	0.23452

Table 2. Sulfur content in the samples

Table 3 shows the density, SG and API values of the crude oil samples from the emulsion samples. Crude oil can be classified as light, medium or heavy, based on its measured API gravity. Light crude is any crude oil having an API value greater than 31.1°. Medium crude oil is defined as any crude that is having API gravity between 22.3° and 31.1°, while heavy crude oil is defined as one that has API gravity below 22.3° [17-18]. Based on API categorization, one of the samples is heavy (W002), three are medium (W/003, W/004& W/006) and the other two are Light (W001 & W005). The density and API gravity of the samples ranged from 0.8376 to 0.9339 (g/cm<sup>3</sup>) and 19.56 to 36.93 respectively.

S/N	Sample ID	Density @ 15oC	SG @ 15ºC	API (°)
1	W/001	0.8564	0.859	33.23
2	W/002	0.9339	0.9367	19.56
3	W/003	0.9110	0.9137	23.36
4	W/004	0.9110	0.9137	23.36
5	W/005	0.8376	0.8401	36.93
6	W/006	0.9094	0.9121	23.63

Table 3. Density, SG and API values of crude oil samples

Figure 1. shows the distribution of the density values of crude oil and the emulsion samples for all samples measured. The density of emulsion for all samples were higher than that of the crude oil. This is expected as the density of emulsion is a function of a number of factors such as the BS&W (amount of water present), nature of the oil (API categorization) and temperature.



Figure 1. Variation in the densities of crude oil and emulsion

Figure 2 shows the variation in the viscosities of the crude oil samples at different temperatures. Sample W/004 has the highest viscosity at all temperatures of measurement and this translates to high specific gravity as seen in Table 1. While, sample W/005 has the lowest viscosity and this also resulted in its low specific gravity. From the chart (Fig. 2 and 5), it is observed that the viscosity of the sample reduces with increase in temperature. As the temperature increases there is an increase in the kinetics of the sample as the molecules in the sample gains energy and therefore moves faster. As temperature increases, the molecular interchange that occurs in the liquid samples is comparable to what exists in a gas stream, with the exception that there are significant cohesive forces that take place between the molecules of a liquid unlike in gases <sup>[8]</sup>. The effect of the temperature is to relax the forces holding the molecules together hence increasing the intermolecular interactions. Solids have more cohesive forces than liquids because the molecules of the solid material are well compacted and would require a stronger energy to free the molecules. So, in order of cohesiveness of their molecules, we have solid>liquid>gas.









Figures 3 and 4 explain the plot of natural logarithm of viscosity (ln  $\eta$ ) against the inverse of the absolute temperatures (1/T) in Kelvin. Modeling the Arrhenius equation and comparing it with the Viscosity temperature study, the natural logarithm of viscosity against the reciprocal of absolute temperature in Kelvin was found to be linear.

Using graphical method and the linear least-squares fitting approach (equ 3), Y = mX + C (3)

where: Y is  $\ln \eta$  (natural logarithm of the viscosity); m is the slope of the straight line graph (which is Ea/R, from the equ 2); X is the reciprocal of the absolute temperature (1/T); C is the intercept of the straight line graph which is the pre-exponential factor, A (see equ 2).



Figure 4. In ŋ against 1/T for samples W/001, W005 & W/006

Applying equations 2 and 3, the activation energies (Ea) for samples W/001, W002, W003, W004, W005 and W006 were found to be 33.56J/mol, 64.06J/mol, 47.78J/mol, 70.54J/mol, 26.61J/mol and 59.90 J/mol respectively. The Entropic factor/Pre-exponential constant (A) for W/001, W/002, W/003, W/004, W/005 and W/006 were found to be 3.48cP, 6.20cP, 2.44cP, 12.51cP, 2.59cP, 3.40 cP respectively.

According to previous report <sup>[8]</sup>, for most liquid systems, the plot of natural logarithm of viscosity versus the reciprocal of absolute temperature in Kelvin is usually linear. The establishment of a linear/straight line graph culminating into good R<sup>2</sup> (regression coefficient) values, shows the correctness of the results obtained. The results obtained show the validity of the relationship between the Arrhenius equation parameters. Consequently, the results obey the linear Arrhenius behaviour.

Figure 5 shows the plot of the kinematic viscosity at different temperatures ranging from 40°C to 80°C of the crude oil samples extracted from the emulsions. The plot followed the same trend as that of the emulsion samples (the viscosity decreases as the temperature increases).



Figure 5. Plot of temperature (°C) against kinematic viscosity (cSt) for the crude oil



Figure 6. Crude oil-emulsion variation at 40°C-80°C

Figure 6. shows the viscosity distribution of the extracted crude oil and the crude oil emulsions for samples W/001 to W/006. A number of factors accounts for the viscosity of crude oil emulsions. Emulsion viscosity is a function of the viscosity of crude oil, viscosity of water, BS &W (amount of water present), API of the crude oil and temperature of measurement. That is;  $\mu_{e} = f(\mu_{o}, \mu_{W,APIo}, T)$  (4)

where:  $\mu_e$  = viscosity of emulsion;  $\mu_o$  = viscosity of emulsion;  $\mu_w$  = viscosity of emulsion; API<sup>o</sup> = API of the crude oil; T = temperature.

The viscosity of emulsion for all samples measured and at all test temperatures should be higher than that of the pure crude oil samples. This is clearly shown in Figure 6. The percent of water present in the emulsion has a strong influence on the viscosity and density of the emulsion as seen in Tables 1 and 3 and Figure 6.

# 4. Conclusion

It can be concluded from this study that temperature affects the viscosity behaviour of crude oil emulsions. The viscosity of the sample reduces with an increase in temperature. As the temperature (thermal energy) increases there is an increase in the kinetics of the sample as the molecules in the sample gains energy and they move faster thereby leading to chemical reactions.

For the emulsion and extracted pure crude oil samples, at different temperatures of measurement, there is a decrease in the viscosity as temperature increases. This result corroborates the study done on Palm oil and soybean oil <sup>[8]</sup>. This study shows that viscosities of emulsion were considerably affected by different temperatures as seen in a similar work done<sup>[19]</sup>. Since, the viscosity of the emulsions is significantly affected by temperature causing an inter-molecular movement and breakage of the cohesive forces/interfacial tension between the oil/water interface. The values of two viscosity-Arrhenius parameters such as the activation energy (Ea) and pre-exponential/entropic factor (A) were also determined and they obey the Arrhenius behaviour. Basically, for all of the samples studied, the plot of natural logarithm of viscosity (ln ŋ) against the reciprocal of the absolute temperature in Kelvin (1/T) followed the Arrhenius linear regime <sup>[9]</sup> and gave a good regression coefficient. In summary, crude oil emulsions from Niger Delta region also obey the already established behaviours. Also, the viscosity of crude oil emulsions is a function of number of factors such as the viscosity of crude oil, BS &W (amount of water present), API of the crude oil and temperature of measurement.

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To whom correspondence should be addressed: Dr. Uche John Chukwu, Department of Pure and Industrial Chemistry, University of Port Harcourt, Port Harcourt, Nigeria; <u>e-mail</u>: <u>lydiuche@gmail.com</u>; uche.chukwu@uniport.edu.ng

# Article

Silicon Dioxide Nanoparticles Boasted Gum Arabic as a Natural Polymer for Enhanced Heavy Oil Recovery

Okechukwu C. Ezeh<sup>1,2\*</sup>, Sunday S. Ikiensikimama<sup>1,2</sup> and Onyewuchi Akaranta<sup>1,3</sup>.

<sup>1</sup> World Bank Africa Centre, Centre for Oilfield Chemicals Research, University of Port Harcourt, Nigeria

<sup>2</sup> Department of Petroleum and Gas Engineering, University of Port-Harcourt. Nigeria

<sup>3</sup> Department of Pure and Industrial Chemistry, University of Port-Harcourt. Nigeria

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#### Abstract

Heavy oil is defined as oil with an API gravity ranging from 10° to 22.3°. Heavy oil reservoirs are considered unconventional reservoirs and are more in the world than conventional reservoirs, and they should not be left unattended. Crude oil from the Niger Delta of API 19.03° and Gum Arabic natural polymer from Bauchi State were used for the flooding experiment to determine the incremental recovery after waterflooding. In this study, various concentrations of gum Arabic were used for coreflooding after waterflooding. The concentrations of Gum Arabic used were 1,000ppm, 3,000ppm, 5,000ppm and 10,000ppm. There was little incremental recovery (2%) when these four different concentrations were used for the brine coreflooding experiment. However, when 1,000ppm of Silicon Dioxide Nanoparticles were blended with 1,000ppm of Gum Arabic, an incremental recovery of 10.15% was observed. This incremental recovery using Silicon Dioxide Nanoparticles during the coreflooding with just a concentration of 1,000ppm has shown the potential synergy between Gum Arabic and Nanoparticles in sweeping Heavy Oil Reservoirs in Nigeria. This study goes ahead to state that companies with heavy oil reservoirs can collaborate with our research centre to carry out extensive studies on their heavy crude and production in a cost-effective and safe manner.

Keywords: Gum Arabic; Heavy Oil; Nanotechnology; Polymer Flooding; Silicon Dioxide Nanoparticle.

# 1. Introduction

#### 1.1. Background

Heavy oil is defined as oil having an API of 10°-22.3° and a viscosity of 10–1,000 cP at reservoir conditions <sup>[1]</sup>. Heavy oils are classified as unconventional reservoirs since they are more widely distributed than conventional reservoirs and should not be ignored. Nigeria holds about 37 billion barrels of proven oil reserves as at 2016, ranking 10<sup>th</sup> in the world and accounting for about 2.25% of the world's total oil reserves of 1,650,585,140,000 barrels <sup>[2]</sup>. The Nigerian government currently derives the majority of its oil earnings from cheap/light oil derived from conventional reservoirs. Globally, these reservoirs are quickly depleting <sup>[3]</sup>.

Nigeria has roughly 42 billion barrels of unconventional oil reserves in addition to its 37 billion barrels of proven reserves. This unconventional reservoir contains heavy oil. According to the numbers above, Nigeria's unconventional resources outnumber conventional resources. This is true for most places across the world. As a result, these unconventional reservoirs should not be ignored <sup>[4]</sup>. Heavy oil reserves are estimated to be 3,396,000 billion barrels on a worldwide scale. As previously stated, conventional oil is dwindling, and we must face the problem of developing heavy oil reservoirs to replace the dwindling light or conventional oil <sup>[5]</sup>.

Waterflooding is seen as a low-cost intervention approach for recovering more oil after primary production. This is for obvious reasons, such as water compatibility, availability, and cost, especially in the offshore environment <sup>[6]</sup>. Waterflooding works better in lighter oils with

lower viscosity and higher API° than in heavier oils with higher viscosity and lower API° <sup>[7]</sup>.Viscous fingering is detected during waterflooding a heavy oil reservoir, which is the unstable displacement of a more viscous fluid by a less viscous fluid <sup>[8-9]</sup>.

Heterogeneities in reservoir rocks cause uneven waterflooding in the reservoir. For zones with high permeabilities, they can receive so much water during waterflooding and this will cause early water breakthrough or thief zone in such reservoirs. As a result, the water by-passes the oil, leaving the unswept oil in the reservoir after the waterflooding <sup>[10]</sup>. The Water-Oil-Ratio (WOR) could reach up to 95% in some cases before becoming uneconomical. Be-cause of pressure drawdown, the pressure in the injection well differs from the pressure in the production well. Infill drilling can be used to recover the unswept oil, but it is costly <sup>[6]</sup>.

Chemical Enhanced Oil Recovery is utilized to address the challenges mentioned above. alkaline (A), surfactants (S), polymers (P), and even nanoparticles are employed separately or in combination as AS, AP, SP, or ASP to sweep and recover more residual oil. The polymers in the water enhance the viscosity of the water, lowering the mobility ratio. Polymer flooding can recover an additional 30% of the original oil in place. The total cost of adopting the polymer flooding approach is lower than that of water flooding due to reduced water production and increased oil production. As a result, money will be saved that would have been spent on dealing with excess produced water. The aim of polymer flooding is to lower the mobility ratio by raising the viscosity of the water and lowering the permeability of the formation <sup>[11]</sup>.

For this research, gum Arabic, a natural polymer, and silicon dioxide (SiO<sub>2</sub>) nanoparticles are used.

# 1.2. Gum Arabic (GA)

Gum arabic is the name given to natural gum exudates of hardened sap from Acacia Senegal (Senegalia Senegal) and Acacia Seyal (Vachellia), deciduous trees. Gum Arabic has other acceptable names like Acacia Gum, Arabic Gum, Gum Sudani, Gum Acacia, Acacia, Senegal Gum, Indian Gum, Gum Hashab, Gum Talha, etc. It is an important cash crop for most countries in sub-Saharan Africa, and Nigeria is not an exception. Sudan is the world's leading producer of gum Arabic, which has greatly benefited the country's economy <sup>[12-13]</sup>. Gum Arabic did not derive its name because it was produced in the Middle East; rather, Gum Arabic was transported to Europe from Arab countries via Arabic ports. As a result, the name Gum Arabic was chosen <sup>[14]</sup>.

Gum Arabic is a complex mixture of polysaccharides and glycoproteins (GPs) and is soluble in water. It forms solutions over a wide range of liquids without turning very viscous. It is used in many industries, including food and beverages, with an E number of 414 (E414). Gum Arabic is primarily used as a natural emulsifier. It is also used in other industries like pharmaceuticals, ceramics, printing, textiles, inks, paper, adhesives, cosmetics, paint, glue, chewing gum, photosensitive chemicals, and pyrotechnics <sup>[13, 15-16]</sup>.

As said, GA is a complex mixture of polysaccharides and glycoproteins (GP's) with a pH ranging from near neutral to mild acidity. It exists as a blended calcium, magnesium, and potassium salt of a polysaccharide acid known as Arabic acid. The framework of GA is made up of 1,3-connected-d-galactopyranosyl units. The side chains are made up of two to five 1,3-connected-d-galactopyranosyl units, joined to the primary chain by 1,6-linkages. Both the fundamental and the side chains contain units of a-l-arabinofuranosyl, a-l-rhamnopyranosyl,  $\beta$ -d-glucopyranosyl, and 4-O-methyl- $\beta$ -d-glucopyranosyl, the last two generally as end units. The chemical composition of GA can change with weather conditions, soil conditions, and the age of the tree and location of the tree [17-18].

Gashua *et al.* <sup>[12]</sup> reviewed several papers on GA and concluded that the characterization of gum Arabic using gel permeation chromatography (GPC), coupled to light scattering, refractive index, and UV detectors, has shown that the gum exudates obtained from both species of *A. senegal* and *A. seyal* consist of three main components. The components are arabinogalactan (AG), arabinogalactan-protein (AGP), and glycoprotein (GP), and they differ mostly in molecular size and protein concentration. The AG, AGP, and GP fractions each account for 90%, 10%, and 1% of the total gum, respectively. They have molar weights of 2,250 kg/mol,

1,200 kg/mol, and 200 kg/mol, respectively, and contain 1%, 10%, and 25-50 % proteinaceous material. The amino acid content/composition of the three components differs greatly, with the GP fraction having the least in common with the other two <sup>[12]</sup>.

# 1.3. Nanotechnology

Nanotechnology is the application of nanoparticles characterized by a size ranging between 1 and 100 nm, and it has been used in the oil and gas industry. It is applicable to exploration, drilling, drilling and hydraulic fracture fluids, oilwell cementing, completion, corrosion & scale inhibition, formation evaluation, production, reservoir characterization & management, processing & refining, and enhanced oil recovery <sup>[19-20]</sup>.

Nanotechnology has been around but was conceived as a new emerging technology in the 1980's. Physicist Richard Feynman (1959) in a presentation tagged "There's Plenty of Room at the Bottom" at an American Physical Society meeting at CalTech, presented the ideas and concepts of nanotechnology and nanoscience. Feynman suggested that processes could be more efficient if scientists could control and manipulate individual atoms and molecules. Professor Norio Taniguchi was the first person to use the word nanotechnology when he was exploring ultra precision machining, after over a decade since physicist Richard Feynman gave his talk. By 1981, individual atoms were visible through the use of Scanning Tunneling Microscope (STM), and modern nanotechnology started <sup>[21-22]</sup>.

Nanotechnology is solving most of the challenges that are associated with CEOR, hence its applicability and synergy with CEOR has been harnessed. CEOR is applied to recover both the bypassed and residual oil trapped in the reservoir. Chemicals like polymers, surfactants, and alkaline are used to achieve this individually or in combination. Using nanoparticles is economical because only a small amount of the chemical is required to achieve the result. This is possible because nanoparticles have a large surface area <sup>[23]</sup>.

Equally, nanotechnology has proven to be environmentally friendly and efficient. Other challenges in CEOR, such as in high temperature and high salinity reservoirs, degradation problems are taken care of using nanoparticles <sup>[24]</sup>. Apart from thermal and salinity resistance, nanoparticles can withstand mechanical degradation, high pressures and high shear rates in the reservoir and can be modified in any reservoir system. Nanoparticles have a high surface to volume ratio, which results in enhanced thermal properties <sup>[25-26]</sup>.

Some authors have commended the applicability and usefulness of namomaterials in solving residual oil challenges in heavy and semi-heavy oil reservoirs over the last decade <sup>[27-28]</sup>.

According to <sup>[23]</sup>, aluminum oxide (Al<sub>2</sub>O<sub>3</sub>) and silicon oxide are good EOR agents, as experiments show. They showed that Aluminum Oxide nanoparticle is good for oil recovery when used with distilled water and brine as dispersing agents. From all their experiments also, they found out that the use of ethanol, Silane treated Silicon oxide gave the highest recovery, while hydrophobic silicon oxide in ethanol also gave good results. Their experiments confirmed that aluminum oxide nanoparticles reduced the oil viscosity, while silicon oxide nanoparticles changed the rock wettability and reduced the interfacial tension of water and oil in the presence of ethanol.

Ogolo *et al.*<sup>[29]</sup> investigated the ability of nanoparticles to trap migrating fines in a sandstone formation. This study used distilled water, brine, and ethanol as base fluids. They found out that two main fines trapping mechanisms in sands were found to be pH values of the surrounding fluids and electrostatic forces of adsorption and that Al<sub>2</sub>O<sub>3</sub> had the most capacity to trap fines. Other trapping mechanisms include zeta potential, nanofluid point of zero charge, and surface charge density of nanoparticles.

Different researchers have used nanoparticles with different chemicals and have been able to solve most of the CEOR challenges <sup>[30]</sup>.

The objective of this paper is to harness the usefulness of Silicon Dioxide nanoparticle in blending the abundant Gum Arabic, that was properly processed for effective flooding in a heavy oil reservoir in Niger Delta of Nigeria.

# 2. Materials and methods

# 2.1. Materials

Table 1 shows the list of materials used in this work and their respective descriptions.

Table 1. List of materials and description.

S/N	List of materials	Description
1.	Heavy crude oil of API <sup>o</sup> 19.03	Obtained from Niger Delta
2.	Core plug	Obtained from Niger Delta
3.	Gum arabic (GA)	Purchased from Bauchi State and processed
4.	Silicon dioxide (SiO <sub>2</sub> ) nanoparticle	Manufactured by Sigma Aldrich
5.	Toluene	Used for cleaning of Core Plug
6.	Sodium chloride (NaCl)	Monovalent Ion for Brine preparation
7.	Calcium chloride (CaCl <sub>2</sub> )	Divalent Ion for Brine Preparation

# 2.2. Equipment/apparatus

Table 2 shows the list of equipment/apparatus used in this experiment and their various functionalities. The OFITE® RPT coreflooding schematics and the actual picture are shown.

Table 2 List of equipment and functionalities

S/N	List of equipment	Functionality
1.	OFITE® RPT	Permeability Measurement/Coreflooding
2.	Redwood viscometer	Determination of Heavy Oil Crude Viscosity
3.	Glass viscometer	Determination of Polymer Solution Viscosity
4.	Soxhlet extractor	Cleaning of Core Plug
5.	Desiccator	Drying of Core Plug
6.	Mud balance	Determination of Specific Gravity
7.	Hamilton beach mixer	Mixing of Polymer and Nanoparticle Solutions
8.	pH/ORP meter	Determination of temperature/pH of polymer solution
9.	Thermometer	Determination of Temperature
10.	Electronic weighing balance	To measure weight of sample
11.	Pycnometer of density bottle	Determination of Specific Gravity of Crude Oil
12.	Measuring cylinder	Measuring required quantity of deionized water
13.	B114 Elgastar deioniser	Preparation of deionized water
14.	Stop watch	For Time taking
15.	Spatula	Collection of polymer and nanoparticles samples

# 2.3. Procedures for the preparation of gum arabic powder

The gum Arabic was purchased from Bauchi State. The gum Arabic exudates were handpicked to remove the impurities, and the clean exudates were pulverized with a sledge hammer in a clean, strong sack bag. The gum exudates were ground and sieved with a 250 m sieve initially and then with a 125 m sieve to obtain the desired gum Arabic powder.

# **2.4.** Procedures for the preparation of gum arabic polymer and silicon dioxide nanoparticle solutions

The gum Arabic powder in various quantities was dissolved in deionized water to get various concentrations, ranging from 1,000ppm, 3,000ppm, 5,000ppm, and 10,000ppm. In percentage, these were 0.1%, 0.3%, 0.5% and 1.0% respectively. A plastic bottle was used in all the solution mixing. Deionized water was measured to 500 mL in each bottle. For the 0.1% or 1,000ppm, gum Arabic powder of 0.5g is measured and mixed into the 500ml deionized water, and 1.5g, 2.5g and 5.0g of gum Arabic powder into 500 mL deionized water representing 0.3% or 3,000ppm, 0.5% or 5,000ppm and 1.0% or 10,000ppm respectively. To determine the exact amount of gum Arabic powder, a spatula was used to scoop it into a filter paper, which was then placed on an electronic weighing balance. When making this measurement, all the fans are turned off so that the exact measurement can be used. When done, the solution is mixed vigorously using a Hamilton Beach Mixer. The solution is allowed for 24 hours to allow

for complete hydration before it is used for polymer flooding. For the nanocomposite solution, 1,000 ppm only was prepared using the same procedures. The long period for hydration helps the flooding to be more effective and efficient and ultimately prevents plugging of the core plug representation of what will be done at the field scale

## 2.5. Procedure for determination of crude oil API°

To ascertain the gravity of the crude, a pycnometer or specific gravity bottle was used. weight of empty bottle  $(W_{eb}) = 32g$ ; weight of bottle and crude oil  $(W_{bc}) = 79g$ ; volume of empty bottle  $(V_c) = 50g$ 

 $Specific Gravity(S.G) = \frac{W_{bc} - W_{eb}}{V_c} = \frac{79 - 32}{50} = \frac{47}{50} = 0.94$  $API = \frac{141.5}{S.G} - 131.5 = \frac{141.5}{0.94} - 131.5 = 150.53 - 131.5 = 19.03^{\circ}$ 

## 2.6. Procedure for coreflooding analysis setup

In this experiment, hydrocarbon recovery by using gum Arabic polymer as a tertiary means and its effectiveness in reservoirs in optimizing heavy oil production.

The experiments performed were conducted at room temperature and suitable differential pressures were selected with the assistance of the equipment used. A gradual rise in device pressure suggested one of two things, an empty accumulator and or a drop in core sample permeability.

Since brine was used to attain 100 % saturation of the cores, the primary recovery process was the first method employed. Once no more brine was produced, oil was injected into the core plugs at 1.5cc/min. This technique carried out the initial saturation of the water,  $S_{wi}$ .

Then next was to flood using water, in which water was pumped at 1.5cc/min into the core plugs until no more oil was generated. It would create saturation of the residual oil, Sor. As a tertiary phase polymer prepared was initiated. After the water flooding, they were inserted into the core plug to determine if they had any impact on oil recovery. The effect of the polymer can be ascertained to be positive if additional oil would be produced thereby increasing the recovery factor (RF).



Figure 1. Core flood setup, 1) pump fluid, 2) pump, 3) valves, 4) displacing reservoir fluid, 5) piston to separate the oils, 6) crude oil, 7) NSB, 8) polymer 9) pressure gauge, 10) bypass valve, 11) hassler cell holder with core, 12) sleeve pressure, 13) effluent into test tubes <sup>[31]</sup>

Since no automatic means of calculating recovery, the experiment had to be consciously observed during the whole flood experiment. Effluent fluid samples were taken manually every 3 minutes at the core holder outlet. The samples were used to measure the quantity of oil and

brine contained and were used to calculate saturation and recovery factor. Figure 1 above shows the schematic of a core flood setup

# 3. Results and discussion

From the experiment carried out as shown in the methodology, the specific gravity of the oil is 0.94, and using API° and specific gravity relationship, the API° of the crude oil is 19.03. The viscosity @ 27°C is 86 cP and the viscosity @ 50°C is 18 cP. Table 3 belows shows the property of the crude oil used in the experiment. Table 4 lists the properties of the core plug used in this experiment.

S/N	Property of crude	Value	
1.	Specific Gravity	0.94	
2.	API°	19.03	
3.	Viscosity (cP) @ 27°C	86	
4.	Viscosity (cP) @ 50°C	18	

Table 3. Properties of crude oil

Table 1	Droportion	of come	~ l ~	weed
Table 4.	Properties	or core	plug	useu

Polymer	L(cm)	D(cm)	BV	PV	θ	K(md)
GA	3.5	3.7	37.65	7.5	0.20	1994

Figure 2 is plotted in order to facilitate understanding on the oil recovery efficiency of waterflooding and gun Arabic polymer at four various concentrations of 1,000ppm, 3,000ppm, 5,000ppm and 10,000ppm, while Figure 3 is plotted in order to facilitate understanding on the oil recovery efficiency of waterflooding with gum arabic nanocomposite. From Figure 2, the waterflooding was able to recover 26.67% of OOIP. The plot is also evident that though four different concentrations of GA of 1,000ppm, 3,000ppm, 5,000ppm and 10,000pm were used for the flooding only additional 2% of oil recovery was recovered. The different concentrations are indicated with different colors on the plot. From Figure 3, when GA of 1,000ppm and SiO<sub>2</sub> of 1,000ppm were used for the flooding, an additional 10.15% of oil was recovered. The SiO<sub>2</sub> nanoparticles have enhanced the natural GA polymer by further altering the wettability of the core plug and reduced IFT.







Figure 3. Recovery efficiency

#### 4. Conclusion

In this polymer flooding study, it is concluded that nanoparticles blended with gum Arabic can enhance the heavy oil from the Niger Delta area of Nigeria. Based on the experimental analysis, the following are evident. Water flooding was able to recover the heavy oil reservoir up to 26.67%. GA natural polymer at four different concentrations was able to recover only 2.0% incremental recovery. GA+SiO<sub>2</sub> recovered an extra 10.15% of heavy oil. SiO<sub>2</sub> nanoparticles are capable of enhancing the abundant GA in Nigeria to recover heavy oil. SiO<sub>2</sub> has obviously improved the recovery above GA alone due to wettability alteration and reduction of IFT.

#### Symbols/Nomenclature

Α	Alkaline
AG	Arabinogalactan
AGP	Arabinogalactan protein
AP	Alkaline-Polymer
API	American Petroleum Institute
AS	Alkaline-Surfactant
ASP	Alkaline-Surfactant-Polymer
BV	Bulk Volume
CalTech	California Institute of Technology
cP	Centipoise
cEOR	Chemical Enhanced Oil Recovery
D	Diameter
EOR	Enhanced Oil Recovery
GP	Glycoprotein
GPC	Gel Permeation Chromatography
IFT	Interfacial Tension
K	Permeability
Ka/mol	Kilogram per Mole
L	Lenath
OFITE® RPT	OFITE® Reservoir Permeability Tester
OOIP	Original Oil in Place
θ	Porosity
P	Polymers
рН	potential of hydrogen
PPM	Parts Per Million
PV	Pore Volume
S	Recovery Factor
S	Surfactants
SP	Surfactant Polymer
Soi	Initial Oil Saturation
Sor	Residual Oil Saturation
STM	Scanning Tunneling Microscope
UNCTAD	United Nations Conference on Trade and Development
UV	Ultraviolent
Vc	Volume of pycnometer
W <sub>bc</sub>	Weight of pycnometer and crude
W <sub>eb</sub>	Weight of empty pycnometer

#### **Declaration of competing interests**

The authors have declared that no competing interests exist in this paper.

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To whom correspondence should be addressed: Dr. Okechukuvu C. Ezeh, World Bank Africa Centre, Centre for Oilfield Chemicals Research, University of Port Harcourt, Nigeria; <u>e-mail</u>: <u>okechukwuezeh@gmail.com</u>

# Article

# **Open Access**

Performance Evaluation and Optimization of Advanced Cyclone System for Palm Waste Induced Flue Gas

Naresh K. Uthama Kumaran<sup>1</sup>, Ho Cheng How<sup>2\*</sup>, Girma T. Chala<sup>3\*</sup>, Poon Han Wong<sup>1</sup>, Edmund De Alwis<sup>1</sup>

<sup>3</sup> International College of Engineering and Management, P.O. Box 2511, C.P.O Seeb 111, Muscat, Oman

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#### Abstract

The performance of a cyclone is primarily determined by three key parameters; namely inlet velocity, pressure drop across inlet and outlet, and particle collection efficiency. To achieve optimal design and development cost, it is vital to have an accurate prediction of pressure drop and particle collection efficiency of the cyclone in response to the variation of inlet velocities. The present study focuses on the design of a serial-configured multi-cyclones system that is capable of reducing the particle emission of the flue gas from 4000 mg/Nm<sup>3</sup> to 150 mg/Nm<sup>3</sup> in a palm waste-fueled boiler system. Optimization of inlet velocity on individual cyclones is performed within the serial-configured multi-cyclones system using a combined computational fluid dynamic simulation (CFD) and the Stairmands's high-efficiency cyclone model-based design analysis. To further improve the accuracy of the design analysis and the CFD simulation, the flue gas at the inlet of the serial-configured multi-cyclones system was sampled, and the distribution of particle size was analyzed. It was observed that an inlet velocity of 20 m/s was found to be optimum, which was later used to determine the total number of cyclones in the design of the serial-configured multi-cyclones system. The use of the newly designed outlet ducting of rectangular shape produces the desired pressure gradient with the presence of negative draft along the outlet ducting.

Keywords: Cyclone; Palm waste; Flue gas; Performance; Optimization.

#### 1. Introduction

Gas or liquid flow separation by utilizing a method of centrifugal force is best achieved by the usage of devices such as cyclones <sup>[1-2]</sup>. This inertial-based separating device is used as the method of pollution control in industries <sup>[3]</sup>, and it is chosen amongst other methods for dust removal mainly due to its high efficiency performance and its cost effectiveness <sup>[4]</sup>. In a cyclone, particles-contaminated air is fed into a chamber of the cyclone. The chamber allows the formation of a spinning vortex that exhausts the clean gas and leaves behind the solid particles to be dispersed along its wall <sup>[5]</sup>. While the lighter weight particles have the tendency to get attracted to the spinning vortex, and travel upwards to the outlet of the cyclone, the heavier particles continue their path towards the dust collector at the bottom of the cyclone <sup>[6]</sup>.

There are many factors affecting the performance of the cyclone; among which the velocity of the particles that travelled along the wall of the cyclone <sup>[7]</sup>, and the particles residence time are the dominant factors <sup>[8]</sup>. The particle residence time defines the time required for the particle to flow through the length of the cyclone to the dust collector <sup>[9]</sup>. The residence time affects the performance of the cyclone as the slower the time the lesser is the efficiency. In fact, roughness of the wall of the chamber, particle density, and inlet velocity are also found to affect the particle residence time <sup>[10]</sup>.

<sup>&</sup>lt;sup>1</sup> Boilermech Sdn. Bhd., Lot 875, Jalan Subang 8, Taman Perindustrian Subang, 47620 Subang Jaya, Selangor, Malaysia

<sup>&</sup>lt;sup>2</sup> Department of Mechanical. Materials & Manufacturing Engineering, University of Nottingham Malaysia, Jalan Broga 43500 Semenyih, Selangor, Malaysia

The performance of the cyclone can be analyzed by the amount of energy used for the movement of the particles within the cyclone, as well as based on its efficiency curve <sup>[11]</sup>. It could be seen that the centrifugal force is directly dependent on the increase of inlet velocity. Higher inlet velocity of the cyclone provides a higher chance for the particles to exit the cyclone <sup>[12]</sup>. Pressure drop of the cyclone is another parameter that has dependency on the change in inlet velocity. It was reported that the increase in inlet velocity resulted in an exponential increase in pressure drop, as well as an increase in the collection efficiency of the cyclone <sup>[13-14]</sup>. It is important to note that the increase in inlet velocity requires less cyclones in a multi-cyclones system to accommodate the gas flow rate.

Though there are many research available on the cyclone, there are limited studies when it comes to the cyclones system for palm waste-induced flue gas, especially for the advanced serial-configured multi-cyclones system. Such an investigative study is even more important with recent tightening of the particle emission limit from 400mg/Nm<sup>3</sup> to 150mg/Nm<sup>3</sup> by the Malaysian Department of Environment. As a measure to fulfill the requirement from the regulatory body, as well as to commit to a cleaner environment, the present study focuses on the performance investigation and optimization of the design of the advanced multi-cyclones system using computational fluid dynamic simulation. To further improve the accuracy of the simulation, the flue gas at the inlet of the multi-cyclones system was sampled, and the distribution of particle size was used to validate the simulation results.

# 2.Methodology

# 2.1. Mathematical model

There are various models used in the design of cyclone, such as Barth *et al.* <sup>[15]</sup>, Leith and Licht <sup>[16]</sup>, Coker <sup>[17]</sup>, Mothes *et al.* <sup>[18]</sup>, Chen and Shi <sup>[19]</sup> and Moore and McFarland <sup>[20]</sup>. The model adopted for this study is the Stairmand model. This model is widely used in the industry and its design is readily available. There are two types of design for the Stairmand model; namely high efficiency, and high gas rate cyclones. The standard cyclone dimension as per Stairmand high efficiency model was chosen in this case.

The inlet velocities were varied at 15m/s, 17.5m/s 20m/s, 22.5m/s and 25m/s to attain the most optimum result with respect to the pressure drop, collection efficiency, and the total number of cyclones needed for the serial-configured multi-cyclones system. This is done by collecting flue gas samples at the inlet of the serial-configured multi-cyclones system where the number of particles in one cubic meter volume is quantified in terms of mass percentage in various ranges of particle size. Using the scaling factor, the figure of collection efficiency can then be changed to other cyclone sizes based on equation (1) <sup>[21]</sup>.

 $\begin{array}{ll} D_2 = [(DC_2/DC_1)^3 \ x \ Q_1/Q_2 \ x \ \Delta \rho_1/\Delta \rho_2 \ x \ \mu_2/\mu_1]^{1/2} \\ \text{where, } D_2 = \text{ mean diameter of the particle separated in the proposed design, at the chosen separating efficiency; } DC_2 = \text{ diameter of the standard cyclone } = 8 \text{ inches (203mm); } DC_1 = \\ \text{diameter of the proposed cyclone, mm; } Q_1 = \text{ standard flow rate (for high efficiency design = 223 m^3/hour,); } Q_2 = \text{ proposed flow rte, m}^3/hour; \\ \Delta \rho_1 = \text{ solid-fluid density difference in standard conditions; } \\ \Delta \rho_2 = \text{ density difference in the proposed design; } \\ \mu_1 = \text{ test fluid viscosity (air at 1 atm, 200°C) = 0.018 mNs/m}^2; \\ \mu_2 = \text{ viscosity of proposed fluid, } \end{array}$ 

The equation above is used to find the scaling factor. The collection efficiency curve could be shifted according to the intended size from the scaling factor. As for the pressure drop in a cyclone, the formula used to calculate is given in equation (2) <sup>[21]</sup>.

 $\Delta P = \rho_f / 203 \{ u_1^2 [ 1 + 2\varphi^2 (2r_1/r_e-1)] + 2u_2^2 \}$ (2) where  $\Delta P = cyclone pressure drop millibar: <math>\rho_f = aas density ka/m^3$ 

where,  $\Delta P$  = cyclone pressure drop, millibar;  $\rho_f$  = gas density, kg/m<sup>3,</sup>;  $u_1$ = inlet duct velocity, m/s;  $u_2$ = exit duct velocity, m/s;  $r_1$ = radius of circle to which the centerline of the inlet is tangential, m;  $r_e$ = radius of the exit pipe, m, $\phi$ = factor from Figure 1 where  $\Psi$ = parameter in Figure 1, given by  $\Psi$ =F<sub>c</sub> A<sub>s</sub>/A<sub>1</sub>; F<sub>c</sub> = friction factor, taken as 0.005 for gasses; A<sub>s</sub>= surface area of cyclone exposed to spinning fluid, m<sup>2</sup>; A<sub>1</sub>= area of inlet duct, m<sup>2</sup>.



Figure 1. Cyclone pressure drop factor (Gopani and Bhargava<sup>[21]</sup>)

**2.2. CFD simulation** 

mainly created by entry and exit losses <sup>[21]</sup>. Moreover, the friction and kinetic energy losses in the cyclone could also cause pressure drop across the cyclone. Using Stairmand model <sup>[20]</sup>, a benchmark value for the simulation was attained. Even though the theoretical values gained would not be as precise; however, a guideline would be achieved when referred with the simulation results. The number of cyclones required was calculated using simple theory of discharge, based on an inlet flow of 62,000 cubic feet per minute and a constant inlet area of the cyclone.

The pressure drop across the cyclone is

ANSYS-FLUENT workbench is used to carry out the simulation for a single cyclone, and row of 12 serial-configured cyclones with a common inlet and a common outlet ducting. The single cyclone simulation is done with Reynolds Stress Model (RSM) and Discrete Particle Model (DPM); whereas for the serial-configured cyclones, only the RSM is performed <sup>[22</sup>.

Description of the strong swirling gas-solid flow was done by the RSM where anisotropy became the base of building this model. Referring to the experimental results, it was seen that certain characteristics agreed such as the pressure drop and flow characteristics. By ignoring the particle-particle collision, the object is taken as a single particle by DPM. Inaccuracy occurs in DPM only when the particle size diameter was found to be larger than 0.1mm <sup>[23]</sup>. The mass and momentum conservation law are given by:

$$\frac{\frac{\partial(\alpha\rho_f)}{\partial t}}{\frac{\partial(\alpha\rho_f u_f)}{\partial t}} + \Delta \cdot (\alpha\rho_f u_f u_f) = 0$$

where,  $\alpha$ ,  $\rho_f$ ,  $u_f$ ,  $\tau$  and  $S_f$  represent gas phase volume, gas density, gas velocity, viscosity stress tensor and mean force in the computational cells, respectively <sup>[23]</sup>.

To describe the strong swirling gas flow in a cyclone, a Reynolds Stress Turbulence Model was adopted.

$U_{k} \frac{\partial (\vec{u}_{i}' \vec{u}_{j}')}{\partial x_{k}} = D_{ij} + P_{ij} + \varphi_{ij} - \epsilon_{ij}$	(5)
$Dij = -\frac{\partial}{\partial x_k} \left( \frac{\mu_t}{\sigma_k} \frac{\partial u'_i u'_j}{\partial x_k} \right)$	(6)
$P_{ij} = -\rho(\bar{u}'_i \bar{u}'_k \frac{\partial u_j}{\partial x_k} + \bar{u}'_j \bar{u}'_k \frac{\partial u_i}{\partial x_k})$	(7)
$\varphi_{ij} = P(\frac{\partial \bar{u}'_i}{\partial x_j} + \frac{\partial \bar{u}'_j}{\partial x_i})$	(8)
$\epsilon_{ij} = 2\mu \frac{\partial \bar{u}'_i}{\partial x_j} \frac{\partial \bar{u}'_j}{\partial x_i}$	(9)

where the Cartesian coordinates system are represented by i, j and k; the diffusion term, stress generation term, dispersion term and turbulence viscosity are represented by  $D_{ij}$ ,  $P_{ij}$  and  $\varepsilon_{ij}$  respectively, and the velocity fluctuations are represented by u`i, u`j and u`k respectively <sup>[23]</sup>.

The trajectory of a discrete phase particle is predicted by ANSYS-FLUENT via a Lagrangian reference frame whereby the force balance projected on the particle is integrated. The particle force and particle inertia equate to the force balance equation as shown below (Cartesian coordinates x direction)

$$\frac{du_p}{dt} = F_D(u-u_p) + \frac{g_x(p_p-p)}{p_p} + F_x$$

(10)

(3) (4) where the drag force per unit particle mass is  $F_{\rm D}(u - u_{\rm P})$ , u as fluid phase velocity,  $u_{\rm P}$  is the particle velocity <sup>[24]</sup>.

$$F_D = \frac{18\mu}{d_P^2 p_P C_c} \frac{C_D R_c}{24}$$

(11)

The parameters above denote that  $\mu$  is the fluids molecular viscosity,  $\rho$  is the fluid density,  $p_{\rm p}$  is the density of the particle, and  $d_{\rm p}$  is the particle diameter. Re denotes Reynolds number, which is given as below.

$$Re \equiv \frac{pd_p|u_p-u|}{u_p-u|}$$

(12)

The drag coefficient,  $C_D$ , is given as  $C_D = a_1 + \frac{a_2}{Re} + \frac{a_3}{Re^2}$ where  $a_1$ ,  $a_2$ , and  $a_3$  are constants for smooth spherical particles over several ranges of Re.  $C_D = \frac{24}{Re} (1 + b_1 R e^{b_2}) + \frac{b_3 R e}{b_4 + R e}$ (13)where  $b_1 = \exp(2.3288 - 6.4581\phi + 2.4486\phi^2)$  $b_2 = 0.0964 + 0.5565\phi$  $b_3 = \exp(4.905 - 13.8944\phi + 18.4222\phi^2 - 10.2599\phi^3)$  $b_4 = \exp(1.4681 + 12.2584\phi - 20.7322\phi^2 + 15.8855\phi^3)(19.2-6)$ which are taken from Haider and Levenspiel <sup>[24]</sup>. The shape factor,  $\phi$ , is (14)

 $\varphi = \frac{s}{s}$ 

where s stands for the surface area of a sphere with the same volume as the particle and S stands for the actual surface area of the particle.

$$F_D = \frac{18\mu}{d^2 n_P C_A}$$

(15)

The equation is for sub-micron particles, Stokes' drag law. In this case,  $F_{D}$  is defined as the above. The factor C<sub>c</sub> is the Cunningham correction to Stokes' drag law, which is given as follows:

$$C_c = 1 + \frac{2\lambda}{d_p} (1.257 + 0.4e^{-\left(\frac{1.1d_p}{2\lambda}\right)})$$
(16)

where the molecular mean free path is noted by  $\lambda$ .

It is also possible to get a high Mach number drag law by simply altering the Mach number greater than 0.4 or Reynolds number greater than 20. For models that involve discrete phase droplet breakup there is an option for dynamic drag law.

# 2.2.1. Boundary conditions

An inlet velocity boundary was selected at the cyclone inlet. The outflow was set at the outlet boundary, and the no-slip boundary conditions were implied to the other boundaries. All the simulations run with the time step at 1e-04 for the gas flow. The Semi-Implicit Method for Pressure-Linked Equations (SIMPLE) was set for the pressure-velocity coupling algorithm. Second Order Upwind was selected as a discretization for momentum whereas PRESTO! (Pressure Staggered Option) was set for pressure. Second Order Upwind scheme was implied for the turbulent kinetic energy, turbulent dissipation rate and Reynolds stress.

An inlet velocity of 15 m/s, 17.5 m/s, 20 m/s, 22.5 m/s and 25m/s were used, and the gauge pressure was zero with reference to the atmospheric pressure. The flue gas exiting the boiler is of high temperature. Gas density was considered to be  $0.585 \text{ kg/m}^3$ , gas viscosity as  $2.4 \times 10^{-5}$  kg/m.s and operating temperature was taken to be 513K.

As for DPM, the inlet and wall boundary were set to be "reflect" and the gas outlet was set to be "escape" and ash outlet was set to be "trap". Other than that, the solid loading was given the same inlet velocity as the gas flow. The particle shape was assumed to be spherical and the particles were scattered evenly around the inlet boundary. Moreover, the particle chosen was solid ash.

The simulation process started off with the meshing of the inner volume of the single cyclone. The mesh type used was a tetrahedral mesh. A study on three different cell numbers was conducted to justify that the cell number is independent from the pressure drop. Table 1 shows the pressure drop and the percentage difference for different mesh sizes.

Table 1. Pressure drop across cyclone with different mesh size

Pressure drop (Pa)	% Difference
401	
401	
420	4.5
409	
	Pressure drop (Pa) 401 420 409

The simulation results were compared with the mathematical model for all velocities. This acts as a verification factor to validate the simulation results, and thus the optimal velocity was identified to proceed to the serial-configured cyclones.

For the cyclones arranged in the series configuration, the same tests were conducted with higher cell numbers, ranging from 1 million to 10 million cells. Pressure drop for the series configured cyclones is depicted in Table 2. In this case, the pressure drop difference was found to be less than 7%, and this is sufficient to justify the independence of the cell number and pressure drop within the range of this study <sup>[23]</sup>.

Table 2. Pressure drop in series configurated cyclones with different mesh size

Cell number	Pressure drop (Pa)	% Difference
1000000	420	
5000000	452	6.6
10000000	448	

#### 3. Results and discussion

#### 3.1. Mathematical model

The collection efficiency for various ranges of particles size at each inlet velocity was tabulated based on the actual flue gas sample that collected at the inlet of the cyclone, before the total collection efficiency was summarized. Table 3 shows the pressure drop across the inlet and the outlet, and the total collection efficiency of the cyclone for each inlet velocity.

Table 3. Pressure drop and total collection efficiency of the cyclone for each inlet velocity



Figure 2. The effect of inlet velocity on pressure drop and total collection efficiency (based on mathematical model)

To further investigate the effect of inlet velocity, both the pressure drop and the total collection efficiency were normalized, and plotted against the inlet velocity, as shown in Figure 2. The effect of inlet velocity on pressure drop was obviously significant, but not the total collection efficiency. By increasing the inlet velocity from 15m/s to 25m/s, the increment seen in pressure drop was as high as 2.8 times, but the total collection efficiency was merely increased by 5.4%.

## 3.2. CFD simulation for a single cyclone

Based on the CFD simulation, the pressure values at the inlet, outlet, and ash outlet of the cyclone were recorded, and plotted against its inlet velocity in Figure 3. It was noted that the normalized pressure at the inlet, outlet, and ash outlet of the cyclone generally increased with inlet velocity. However, a slightly different trend for ash outlet pressure was observed, whereby the increment of pressure appeared to saturate at the velocity of 22.5m/s.



Figure 3. The effect of inlet velocity on inlet pressure, outlet pressure, and ash outlet pressure (based on CFD simulation)

By plotting the weight distribution of various particles sizes from the actual flue gas sample that collected at the inlet of the cyclone, Figure 4, the removal of particles within the range of 1 $\mu$ m to 12 $\mu$ m deserved higher attention in determining the optimum inlet velocity of the cyclone. Therefore, a set of particles of 1.11 $\mu$ m, 4.06 $\mu$ m, 7.77 $\mu$ m and 10.75 $\mu$ m were selected for the simulation of the particle tracking using DPM, with inlet velocities varied from 15m/s, 17.5m/s, 20m/s, 22.5m/s and 25m/s.





It can be seen from Table 4 that the maximum collection efficiency is achieved for particle size of 10.75µm across all velocities, whereas the minimum efficiency of 79% to 86% was witnessed for particle size of 1.11µm. However, an increasing trend of the efficiency was noticed amongst all the inlet velocities with the increase of particle size, as shown in Figure 5. Higher inlet velocity causes greater centrifugal force which makes the particle move towards the wall faster which in turn increases the collection efficiency. Other than that, the increase in efficiency across the velocities is generally due to the increase in pressure gradient at the cyclone's ash outlet. This results in the formation of vortexes in the cyclone, thus giving rise to higher particle collection efficiency.



Table 4. Collection efficiency of different particle size for different velocities

Figure 5. The effect of inlet velocity on collection efficiency for different particles size

# **3.3. Results comparison between mathematical model and CFD simulation of a single cyclone**

The pressure drop across the inlet and the outlet of the single cyclone that was obtained based on the mathematical model and the CFD simulation was compared. The results are shown in a normalized scale to highlight a similar trend in both cases.



Figure 6. Normalized scale of pressure drop across inlet and outlet of the cyclone

As seen in Figure 6, the difference in both pressure drops is consistent, and the value is increasing with velocity. It can also be seen that the mathematical model values are

consistently higher than the values of CFD simulation. This is mainly due to the reason that only the gas flow was considered in the simulation, whereas the gas flow with the addition of particle tracking was considered in the mathematical model. It is suggested that the dense particles cause the gas flow to have a higher pressure drop <sup>[25]</sup>.



Figure 7. Cyclone efficiency curve

Figure 7 shows the comparison of the collection efficiency curve for the simulated and calculated results at inlet velocity of 20m/s. The reason for taking this inlet velocity as it is the median value of the range of inlet velocities investigated. It is observed that the efficiency increased exponentially with particle size, and the trends appear to be the same for the results that attained in both the calculated and simulated approaches. This proves that the values obtained in simulation are in good agreement with that from the model.

Based on the observation, larger particles are generally heavier, and easier to be collected; whereas smaller and lighter particles are easily escaped through the outlet. Figure 7 shows that both methods show the same trend for  $1.11\mu$ m with a steep drop in the calculated value. This is because for the simulated values, particle collision and wall attrition are not in the consideration [<sup>23]</sup>. Moreover, only a particular particle size is used in the simulation for the entire flow.

Table 5 shows the comparison of pressure drop and collection efficiency using the simulation results. The selection of the optimum inlet velocity requires a low pressure drop for better performance, and high collection efficiency for achieving the intended outcome by using the least number of cyclones for a smaller foot-print of the advanced cyclone system. From these assessments a conclusion was made that the best inlet velocity that accommodates for all the criteria is 20m/s.

Inlet velocity (m/s)	Pressure drop, ΔP (Pa)	Collection efficiency (%) @4.06µm
15	202.94	95.28
17.5	312.71	95.58
20	409.50	97.22
22.5	485.64	97.64
25	25 670.54	

Table 5. Comparison of 3 key parameters for various velocities

# 3.4. CFD simulation for serial-configured cyclones

With a proper understanding of the optimum performance of a single cyclone, CFD simulation of an array of twelve serial-configured cyclones was carried out to optimize the design of the ducting system that consists of a common inlet ducting, and a common outlet ducting. In this case, the velocity of the gas flow in the common inlet ducting was set to 20m/s as it was deducted to be the optimum value for a single cyclone. The results from the simulation were analyzed for various ducting designs, as described in the following cases A, B, and C.



3.4.1. Case A: A common ducting system with no partition at the inlet ducting





# (b)

Figure 8. Case A (b) Side view of the outlet ducting

Figure 8 shows the simulation results of the serial-configured cyclones system in the common ducting system. In Figure 8(a) the distribution of flue gas was uneven in the inlet ducting; whereby the flue gas velocity was found to reduce from 15m/s at the leading edge of the ducting to 5.23m/s at its trailing edge. Figure 8(b) shows the velocity distribution of the serialconfigured cyclones and their common outlet ducting. The velocity distribution in the outlet ducting revealed very poor uniformity, especially for cyclones 1, 2, 3 and 4. This allows the particles to be trapped in the outlet ducting and results in re-entry of these trapped particles into the cyclones. Besides, these trapped particles could also lead to clogging issues which required frequent maintenance work.

Figure 9 shows the inlet velocities of these twelve serial-configured cyclones. The velocities obtained across those cyclones were very high. The pressure drop was also obtained by sub-tracting the pressures at the entry point of the inlet ducting with that of the exit point of the outlet ducting. As a result, the pressure drop was 375.37Pa which was lower than the measured single cyclone value of 419Pa.



Figure 9. Inlet velocities of cyclones (Case A)

# 3.4.2. Case B: A common ducting system with partition at the inlet ducting

In this case, two baffle plates were added to the inlet ducting to segregate the inlet ducting into three zones to reduce the inlet velocity of the cyclones, as illustrated in Figure 10(a). The flow of flue gas diverted to cyclones 1-5 for zone 1, cyclones 6-9 for zone 2, and cyclones 10-12 for zone 3.





Figure 10. Effects of partition at inlet ducting, (a) Top view, (b) Side view and (c) Vortex formation at inlet ducting

The partition caused formation of vortex in all three zones could be seen clearly in Figure 10(c). The vortex causes the particles to be held back in inlet ducting that may cause clogging in the long run. Figure 10(b) reveals the improvement in flow distribution in the outlet ducting as compared to the model in Case A.





As compared with Figure 9 in Case A, Figure 11 for Case B clearly shows significant variation in inlet velocities across all the twelve cyclones. This indicates poor uniformity in performance in these twelve cyclones. For example, the 6<sup>th</sup> cyclone in comparison has a very low inlet velocity due to high vortex formation. The pressure drop for this model was 406.16Pa, which was higher than Case A. However, there were a few problems witnessed in this model such as vortex formation and higher variation of inlet velocities.

# 3.4.3. Case C: A common ducting system with rectangular outlet ducting

Since the inlet ducting was of sufficient flow as evident in Case B, no further optimization was made in the design of the inlet ducting for the Case C. Instead, higher emphasis was focused on the flow of outlet ducting, inlet velocity of cyclones and pressure drop. In this case, the outlet ducting design was modified from a gradually-reduced cross-sectional area to become a uniform rectangular area throughout the whole length of the ducting. Figure 12(a) shows an evenly distributed flow at the inlet ducting. Although zone 2 shows a higher velocity of flue gas, the variation among the zones were not vastly different. This indicates that all the cyclones would be operating around the optimum inlet velocity of 20m/s, as illustrated in Figure 13.







#### (c)

Figure 12. Effects of rectangular outlet ducting, (a) Top view, (b) Side view and (c) Vortex formation for outlet ducting

It can be seen from Figure 12(b) that the flow of inlet velocity is well distributed as the first three cyclones outlets attained an average velocity of 4.23 m/s in the outlet ducting. The trailing edge of the outlet ducting achieved a velocity of 9.97m/s. In Figure 12(c), the formation of vortex at the outlet ducting was very minimal. This shows that rectangular outlet

ducting provides a viable solution as it minimizes the vortex formation problem, and results in more uniformly distributed gas flow.



Figure 13. Inlet velocities of cyclones (Case C)

Based on Figure 13, it is seen that the inlet velocity flow is of the best amongst the other cases. The flow is uniformly distributed without having many peaks or lows. However, cyclones in zone 3 had a higher range of velocity across all cyclones. The pressure drop attained was 380.25Pa.

Figure 14 illustrates the outlet ducting pressure distribution of Case B and Case C. Case B shows a negative draft at the zone around cyclones 8 to 12, while a positive draft is observed at the zone around cyclones 1 to 7, Figure 14(a). The negative draft indicates a natural flow gradient for the gas to exit the outlet ducting. However, the positive draft opposes the gas flow, and a lower pressure gradient prevents the gas from exiting the outlet ducting. Such an issue could be avoided in Case C with the presence of negative draft along the whole outlet ducting, Figure 14(b).



Figure 14. (a) Side view pressure distribution of Case B



(b)

Figure 14. (b) Side view pressure distribution of Case C

## 4. Conclusion

A study on performance evaluation and optimization of advanced cyclones system for palm wasted induced flue gas was performed using both Stairmand's mathematical model and CFD simulation. As the inlet velocity increases, the collection efficiency increases regardless of the particle sizes. While the results obtained from CFD simulation are in good agreement with that of the mathematical model, a slight deviation in values was considered to be due to the assumption of non-collision between particle-to-wall, and particle-to-particle in CFD simulation.

During the simulation of serial-configured cyclones, a high range of inlet velocities was recorded for the inlet ducting design with the absence of partition. Such a high inlet velocity was deemed to be undesired due to potential high erosion wear of the internal wall of the cyclone. With the addition of partition in the design of the inlet ducting, more uniformly distributed flow was observed in the outlet ducting, but the formation of undesired vortex was also evident. The formation of vortex at the outlet ducting was able to minimize with the change of shape of the outlet ducting from tapered to rectangular, by keeping the same design of the inlet ducting with partition. The use of the newly designed outlet ducting of rectangular shape also produces the desired pressure gradient with the presence of negative draft along the outlet ducting.

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To whom correspondence should be addressed: Dr. Girma T. Chala, International College of Engineering and Management, P.O. Box 2511, C.P.O Seeb 111, Muscat, Oman; e-mail: <u>girma@icem.edu.om</u>

# Article

Analysis of Induced Seismicity at Pohang Geothermal Power Plant and Examination of Public Perception following the Incident – A Perspective

Anil Kumar Vinayak, Lianne Lefsrud

Department of Chemical & Materials Engineering, University of Alberta, Canada

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#### Abstract

Alternative sources of energy are the need of the hour and numerous technological innovations have been explored in recent years to facilitate the demand for sustainable energy sources. However, premature deployment without completing a comprehensive risk assessment and formulating mitigative measures can lead to catastrophes that could, in addition to financial losses and human fatalities, tarnish the public perception of an alternative energy source and hinder its proliferation. The Pohang Earthquake aptly demonstrates this phenomenon where Enhanced Geothermal System (EGS), a relatively new technology, led to a large scale induced seismic event. This paper explores the risk posed by fluid injection in the EGS process considering the induced seismic event at Pohang and includes a Bow-tie analysis that proposes preventive and recovery controls to keep the impact of such events to a minimum. The paper also examines the change in Korean public perception to geothermal energy following the induced earthquake.

**Keywords**: Sustainable Energy; Premature deployment; Risk assessment; EGS; Induced seismicity; Bow- tie; Public perception.

#### 1. Introduction

Energy sources have gone through numerous transformations over the years. From wood and coal to petroleum and natural gas, these transitions were instigated based on certain factors such as availability, accessibility, energy density, efficiency, cost of extraction and industrial as well as domestic demand <sup>[1-2]</sup>. At the turn of the century, a new facet has been included into this evolving chain – sustainability <sup>[3]</sup>. Sustainable energy extraction and generation featuring alternative fuels sources have become the crux of most environmental policies and receive substantial attention from the public. The perception of these alternative or renewable sources is generally favorable, however, the status quo can change immediately following an adverse incident or the discovery of a high-impact potential risk. Nuclear Energy and the Chernobyl disaster depicts this phenomenon quite vividly.

Geothermal Energy has been at the forefront of sustainable development projects and policies and the attention it receives is substantiated by the numerous benefits that this source possesses. The advantages of geothermal energy include small carbon footprint, natural replenishment, increased stability, and reliability <sup>[4]</sup>. Both government and private sectors are keen to tap into this plentiful alternative source with projections indicating a 28% growth in renewable energy extraction and helmed by Asia for the most part <sup>[5]</sup>. Despite the appealing nature of these benefits, a closer look will reveal that the extraction process comes with its own set of caveats. The most concerning among these is the potential for induced seismicity. Geothermal energy extraction has been known to trigger microseismic activity and the repercussions have often been sidelined due to the dearth of significant incidents. The dynamic changed following the aftermath of the 2017 Pohang Earthquake in South Korea. The event was measured to be of magnitude 5.5 on the Richter Scale and caused substantial economic damage. Further investigation and analysis revealed that the Pohang Enhanced Geothermal System (EGS) project was the root cause of the incident and acted as the trigger for the earthquake <sup>[5]</sup>. There is much that is not known regarding EGS technology, primarily because the technology is relatively new. Risk Assessment and Environmental Impact studies for the most part have been superficial and sufficient base data is not presently available to perform comprehensive analysis.

The Pohang event is an acute example of oversight by the decision makers and serves as the perfect backdrop to motivate change <sup>[6]</sup>. Government policy makers who sanctioned the EGS project should have delayed the approval until 3-tier geographical analysis was completed <sup>[7]</sup>. Moreover, a small-scale pilot plant should have been set up first to understand the long-term repercussions of EGS prior to industrial-scale deployment. The Pohang incident serves as a wake-up call for decision makers in the government focused on sustainability projects involving nascent technology. A committee consisting of discipline experts as well as engineering professionals need to perform extensive site surveys prior to giving the nod to the decision makers <sup>[7]</sup>. This is definitely a time-intensive process, however, the potential damage incurred by a significant induced seismic event is considerable which justifies the time delay.

This paper explores the risk of induced seismicity brought about by Enhanced Geothermal Systems and the changes in public perception following the event. The paper will further discuss the risks posed by EGS in context of the Pohang project and suggest mitigation strategies to minimize the risk.

#### 2. Induced seismicity

Seismicity is defined as the measure of the frequency and distribution of earthquakes in a particular geographical location in relation to its strength or magnitude. It is calculated using the following formula:

 $Seismicity = \frac{\sum_{i} E_{soi}}{\delta \phi_o \delta \lambda_0 \delta h_0 \delta t_o}$ 

(1)

(3)

where:  $E_{s0i}$ : energy of a single seismic event;  $\delta \phi_0$ : latitude interval;  $\delta \lambda_0$ : longitude interval;  $\delta h_0$ : hypocenter interval;  $\delta t_0$ : seismic event time interval.

Induced seismicity refers to the seismic events or activities that are caused because of human activity or interference. Induced seismicity can also be defined by the equation above <sup>[8]</sup>.

# 2.1. Mechanism

For induced seismic activity to occur, three conditions must be met at the extraction zone where the rock blocks are situated <sup>[9]</sup>. These include,

- The presence of a pre-existing fault at the site of origin.
- The fault is critically stressed and susceptible to slip
- The inducing well should be capable of inciting the slip

The operation that leads to fault failure is often fluid injection. There are two primary mechanisms that govern this mode of failure, pore pressure diffusion and increment of rock stress. Among the two, the former has a more pronounced effect. Pore structures in the rocks lead to stress transmission owing to the injection of fluid and the time lag between filling and inception of seismic activity can be used to confirm induced seismicity <sup>[9,11]</sup>. This time lag as well as the pressure diffusion can be described using Darcy's Law & the Seepage Equation in 2-D described below.

$$v_x = k \frac{\partial_p}{\partial_x}; \ v_y = k \frac{\partial_p}{\partial_y}$$
(2)

where: k: permeability constant; p: pore pressure.

$$\frac{\partial p}{\partial p} - c \frac{\partial^2 p}{\partial p}$$

 $\frac{\partial t}{\partial t} = c \frac{\partial^2 z}{\partial^2 z}$ where: t: time; c: coefficient of consolidation.

The pressure diffusion ultimately leads to fracture growth which in turn results in a slip, thereby triggering a microseismic event or earthquake. Therefore, determining the rate of growth fracture with respect to the fluid injection time will help in assessing the likelihood and

creating a timeline for the seismic activity <sup>[12]</sup>. The fracture growth can be assessed using the following equation:

$$r_f(t) = \frac{g_1 t}{2h_f w + 4h_f C_L \sqrt{2t}}$$

(4)

where:  $r_f$ : fracture half-length as a fraction of injection time; t: injection time;  $q_1$ : injected fluid rate (average);  $C_L$ : fluid-loss coefficient.

The three equations mentioned above (eqn. 2–4) illustrate the mechanism of induced seismicity and can be used to prove the same with respect to pore size and pore pressure changes.

#### 2.2. Enhanced geothermal system

One of the major contributors to induced seismicity during geothermal energy extraction is EGS. As the name suggests, EGS involves an artificially created reservoir. It is employed when the hot rocks in the chosen extraction site offer minimal permeability as well as fluid saturation. This scenario poses a host of challenges as fluid, permeability and heat are critical for the success of a geothermal operation. Therefore, EGS involves the injection of a fluid into a section of the rock structure with a pre-existing fracture. The stress derived from fluid injection causes these fractures to open and subsequently enhances permeability <sup>[13]</sup>. This makes the reservoir more potent in terms of both yield and ROI potential.

EGS is a relatively young technology system and is most popular in Iceland, Australia, United States of America, and Germany <sup>[13]</sup>. In addition to minimal greenhouse emissions, EGS has the potential to bridge the geographical limitations that tend to hold back the proliferation of geothermal energy. However, being a nascent technology, the risks associated with EGS are not fully understood and new evidence suggests that the process could very well disrupt fault lines leading to seismic events with drastic consequences <sup>[14]</sup>. This discussion on induced seismicity reached its crescendo following the Pohang earthquake, the largest recorded man-made earthquake!

#### 3. Incident analysis

#### 3.1 Overview

On 15th of November 2017, the city of Pohang in the North Gyeongsang province of South Korea was rocked by an earthquake measuring 5.4 on the Richter scale. The mainshock epicenter of the earthquake was found to originate from the EGS site of the Pohang Basin. Foreshocks and aftershocks were detected in the radius of 0.6 - 2.5 km from the epicenter <sup>[15]</sup>. In terms of scale, the Pohang earthquake was the second largest seismic activity in the recorded history of South Korea. A total of 90 injuries were reported following the earthquake and the property damage was estimated by various sources to be in excess of USD 52 million <sup>[16]</sup>.

#### **3.2 Seismic history of South Korea**

South Korea is not geographically located on any major tectonic plate line and is classified by geologists to be a Stable Continental Region (SCR) <sup>[17]</sup>. The Korean peninsula is primarily influenced by the Eurasian and Indo-Australian plates. However, it is located more than eight hundred kilometers away from the closest plate boundary and is consequently not prone to any major seismic activity exceeding magnitude of 5 <sup>[15]</sup>.

From the evaluation of the tectonic map depicting Subduction Zones of Northeast Asia utilized by Kim et al. in their study of the Pohang earthquake, it can be deduced that the likelihood of natural earthquakes is minimal and South Korea should be safe from catastrophic seismic events <sup>[15]</sup>. A comprehensive list of large-scale seismic events that occurred in South Korea is summarized in the Table 1.

From the Table 1, it can be surmised that the Pohang earthquake was an anomaly and that the frequency of earthquakes of magnitude of 5 greater have increased in the past decade.

Name	Date	Time (Kst)	Magnitude (M <sub>I</sub> )	Location
Sangju earthquake	09/16/1978	17:07:06	5.2	Sangju, North Gyeongsang
Hongseong earth- quake	10/07/1978	09:19:52	5.0	Hongseong County, South Chungcheong
Ongjin earthquake	03/30/2003	11:10:53	5.0	Ongjin County, Incheon
Uljin earthquake	05/29/2004	10:14:24	5.2	Uljin County, North Gyeong- sang
Taean earthquake	04/01/2014	19:48:35	5.1	Taean County, South Chung- cheong
Ulsan earthquake	07/05/2016	11:33:03	5.0	Dong District, Ulsan
Gyeongju earth- quake	09/12/2016	11:32:54	5.8	Gyeongju, North Gyeongsang
Pohang earth- quake	11/15/20 17	05:29:32	5.4	Buk-gu, Pohang, North Gyeongsang

Table 1. List of major seismic events in South Korea since 1976 [18]

## 3.3. Human-induced event

From the previous section and the proximity of the earthquake's epicenter to the EGS site, it can be concluded that the earthquake was not caused due to plate tectonics. During the EGS process, Hydraulic Stimulation via fluid injection was carried out in four separate phases. A study conducted by Kwang-Hee Kim et al., revealed that seismic activity in the region peaked following a few days after each injection and subsequently died down <sup>[15]</sup>. Moreover, the study was able to establish a correlation between the quantity of fluid injected and the magnitude of the micro-seismic activity. All the evidence points to a slip occurring in a pre-existing fault line. Therefore, it can be conclusively stated that the Pohang earthquake was a human-induced event.

# 3.4. Results of analysis

The Pohang incident acutely depicts a disaster that was the result of unchecked accelerated innovation. The technology used was robust, however, the repercussions following its application were not fully understood and deployed prematurely. This shows the need for comprehensive risk assessment and preliminary studies prior to sanctioning projects, particularly in the field of energy extraction.

#### 4. Risk management

The Pohang earthquake highlights the threat of induced seismicity and shows the importance of hazard identification and risk management. There are numerous methods to perform risk management with each of them having their own set of advantages and disadvantages. A Bow-tie analysis is carried out in this paper. Bow-tie analysis is a powerful risk evaluation tool that can be used to visually represent all possible combinations of risk, threats and consequences of an identified hazard along with the respective preventive as well as recovery controls. The Bow-tie method is perfect to gain an overview of the associated risks in a process and can significantly help the risk management team in establishing mitigating protocols <sup>[19]</sup>.

# 4.1 Assumptions

Prior to executing the analysis, certain assumptions need to be made to ensure the integrity of the process and lend precision to the exercise. The assumption for conducting the risk analysis are as follows:

- The EGS site is considered as the epicenter of the earthquake and the plant will be assumed to be a single collective unit.
- The presence of a pre-existing faultline prior to the earthquake serves as the base for all analysis.
- The influence of plate tectonics is negligible.
- Seismic activity occurring in the past are not included in the scope of the analysis.
- Only fluid injection is assumed to induce seismic activity and the influence of other processes are not considered.
- A closed system is assumed to identify the conjectural solution free from external influence.

#### 4.2. Bow-tie analysis

With the Pohang earthquake as the basis, a Bow-tie analysis of EGS via fluid injection is performed below (Figure 1) with top event being induced seismic activity. The threats and preventive controls are indicated on the left of the Bow-tie while the right side displays the consequences along with the recovery controls. The detailed description of each segment is provided in Table 2.



Figure 1. Bow-tie for induced seismic activity

Table 2. Description of preventive and recovery controls [7,12,17,20-22]

#	Preventive Control	Description	#	Recovery Control	Description
a.	Nucleation Model Forecast- ing	Use hydrologic and fluid injection data to forecast induced seismic- ity by estimating the intensity of pore pressure diffusion. This will help to determine the rate of pro- gress thereby allowing for miti- gation via modification of injec- tion timelines.	j.	Moment Re- sisting Framework	Structural design of buildings on- site is resistant to bending mo- ment and reduce likelihood of cav- ing in during seismic activity.
b.	Numerical Sim- ulation	Employ a 3D model for fluid flow by incorporating the hydraulic properties of the injection fluid and subsequently estimate the epicenter for a seismic event. In- jection zones will be relocated farther away from the probable epicenter.	k.	Reinforced Modular De- sign	Structures are made of modules that can be easily modified or re- inforced. Structural collapse does not upend modular integrity and can be easily reassembled.

#	Preventive Control	Description	#	Recovery Control	Description
c.	3D in-situ stress model- ling	Estimate the stress field around the extraction site to determine the critical points at which fault expansion will occur. Extraction parameters are modified accord- ingly to minimize expansion rates.	Ι.	Robust Evacuation Plan	A comprehensive plan can in- crease the efficiency and pace of personnel evacuation from the danger zone during a seismic event.
d.	Geological Sur- vey	Understanding the geological nu- ances of the site allows for de- tailed mapping of the faultlines. This information could be used to mitigate fault expansions by identifying injection sites that re- sult in minimal expansion.	m.	Disaster Re- sponse Pro- tocol	On-site personnel trained in emer- gency protocols can react more ef- ficiently during an incident and, thereby, reduce casualties.
e.	Field Mapping	Geospatial data can be used to generate models that predict the likelihood for slip. The results will help in outlining restorative pro- tocols.	n.	Control / Isolate Igni- tion Sources	Control ignition sources such as static electricity (earthing) and store flammable gases and liquids away from central locations on site with adequate clearance space.
f.	Monitoring Causative Stress	Monitoring zones of the geologi- cal formation with stress deform- ities over extended periods will aid in the isolation of regions where stress caused by injection becomes greater than the rock's strength.	ο.	Pressure Re- lease Valves	Automated pressure release valves will help to prevent anom- alies such as pressure build-up caused by seismic activity.
g.	Use Low Dam- age Drilling Fluid	The composition and properties of the fluid can play a crucial role in exacerbating stress fractures. Switching to a more compatible fluid can lessen the stress.	p.	Fire Retard- ants	Employ automated dispersion of foam-based fire retardants and sprinklers to deter propagation of a fire.
h.	Increase delay times between enhancements	Multiple hydraulic enhancements are a necessity. Extending the downtime between each injection phase will prevent the regional accumulation of stress.	q.	Central Fire Handling	Central monitoring station to han- dle fire related events during an incident and coordinate fire quenching efforts to minimize spread.
i.	Reduce Inten- sity of En- hancement	Flow rate of fluid during injection is maintained below the stress failure threshold.	r.	Failure Re- lease Sys- tem	Controlled venting of toxic gases using washers to enable auto- mated release during seismic ac- tivity.

# 4.3. Results

The Bow-tie analysis indicates the dire consequences of an induced seismic event and reveals the importance of 3D mapping prior to fluid injection. Mitigative measures in this scenario revolve around selecting the most conducive injection site for enhancements and the intensity of the enhancement with the type of injection fluid playing a supplementary role. Recent research has indicated that supercritical carbon dioxide can be used in place of wastewater, the typical injection fluid <sup>[23]</sup>. The physical and chemical interactions of carbon dioxide with rocks in the reservoir is not as pronounced as water and limits pore pressure diffusion and stress factors. Injecting carbon dioxide in zones that are not directly passing through fault lines will help to prevent causative shear and stress, thereby effectively reducing the likelihood of slip <sup>[23]</sup>.

The potential of EGS is not yet fully realized and the fledgling technology is ripe for innovation. However, the execution of the concept requires detailed geological study to ensure that the enhancements do not accumulate towards a large-scale seismic event as observed in Pohang.

### 5. Public perception of geothermal energy

Climate change is an imminent threat that has already begun to spell disaster in certain parts of the world. Naturally, the interest in alternative energy capable of supporting a sustainable future has exponentially increased of late. Geothermal energy falls under this spectrum and is viewed as a viable energy source in many parts of the world. Unfortunately, the Pohang earthquake had a marked effect on the public perception of geothermal energy. This is synonymous to the public opinion of nuclear energy following the Chernobyl disaster. This section examines the public perception of geothermal energy in South Korea considering the induced seismic event at Pohang.

# 5.1 South Korean energy policy

Much of the Korean public's opinion of geothermal energy stems from the country's energy policy. As per the latest long-term plan, Korea seeks to transition to renewable energy by sourcing 35% of its energy supply from alternative sources by 2040. This is a Herculean task given that in 2018, 85% of the country's energy requirement was sourced via fossil fuels <sup>[24]</sup>. The influence of the policy extended to numerous primary and secondary sectors which ultimately spurred on a renewable revolution in the minds of the public. With EGS based geothermal energy being touted as the game-changing innovation spurring the paradigm shift, the technology was welcomed with open arms by Koreans. However, the major caveat is that risk perception was lacking amongst the public leading to creation of premature point of views.

Energy policies take into consideration both economic and technological factors, however, their execution involves various other limiting factors as well. They are highly influenced by external influences such as the political climate, international commitments, and logistical constraints. A study conducted by Woo et al. was able to establish a causal link between the Pohang earthquake and EGS project <sup>[25]</sup>. Therefore, it can be inferred that the technology was deployed prior to establishing adequate parameters for mitigating seismic activity caused by the fuel injection process. The South Korean energy policy is formulated around the 2040 deadline which results in energy projects being sanctioned at a greater pace and, as is the case with the Pohang incident, without due diligence <sup>[24]</sup>.

# 5.2. Induced seismicity – EGS and the Pohang earthquake

As described in section 3 earlier, it can be conclusively established that the Pohang Earthquake was not a natural occurrence and was a human-induced event. Woo et al. were able to conclusively establish the causal link between EGS and the Pohang Earthquake <sup>[25]</sup>. They performed a parameter analysis involving hypocenter determination, initial and relative locations, magnitude determination, earthquake size distribution, focal mechanism solution, and hierarchical clustering analysis. The results were compared with hydraulic experiments and spatial distribution data to establish the relation between hydraulic stimulations at the EGS site and earthquake sequences <sup>[25]</sup>.

# 5.3. Social consequences

The investigation shown in section 5 was able to conclusively establish that the Pohang Earthquake was caused by the activities of the EGS plant. The Geothermal Energy consortium overseeing the plant faced severe backlash from the Pohang community once the report went public <sup>[26]</sup>. Social media and online bulletin boards buoyed up the issue while support groups were formed for those who suffered physical, emotional, and financial losses due to the incident. The media broadcasted the aftermath of the earthquake in tandem with the induced seismicity report and the fall in grace of the EGS Geothermal plant was imminent.

The social consequences of EGS based geothermal energy extraction and the drastic shift in public perception can be attributed to the psychosocial cognitive effect known as anchoring <sup>[26]</sup>. Often considered a bias, anchoring refers to the decision-making that is concentrically elaborated from the initial piece of information <sup>[26]</sup>. This serves as the basis or anchor for all subsequent decisions with judgements being tailored to align with the anchor. For the Pohang incident, establishing the causation between EGS process and induced seismicity let down the anchor that led to a dip in the favorability of geothermal energy where the public perception was disproportionately skewed against geothermal energy <sup>[27]</sup>. A study conducted by Dong-Hyeon Im *et al.* to evaluate the favorability of various energy sources among Pohang residents following the induced seismic event revealed certain interesting points pertaining to the social consequences <sup>[26]</sup>.



Figure 2. Favorability of energy sources ranked by Pohang residents following the earthquake with 1 being the least favorable and 5 being the most  $^{[26]}$ 

From Figure 2, it is apparent that Pohang Residents were extremely averse to the use of geothermal energy following the Pohang earthquake. It should be noted that geothermal energy was ranked below nuclear and coal, the energy sources typically vilified by media houses and green policy makers. Moreover, Pohang residents are well-versed with the concept of EGS which makes the change in perception more than a knee jerk reaction. The final nail in the coffin for the EGS plant in Pohang was the financial losses left in the wake of the disaster. An estimated \$52 million worth of property and infrastructure damage was initially reported <sup>[16]</sup>. This figure does not include the privately owned losses and the harm caused to mental health as a result. Majority of the losses were due to structural collapses and could have been avoided if the recovery control shown in the Bow-tie analysis, particularly reinforced modular design and moment resisting framework, were employed to construct these structures <sup>[28]</sup>.

#### 5.4. Aftershock of perception shift

The polar shift in perception following a disaster has far-reaching implications. Once the EGS plant was confirmed to be the trigger for the earthquake in 2018, public outcry put an immediate stop to its operations. Risk mitigation teams were deployed to assess strategies to ensure safer operations while EGS expansion plans across the region were put on hold. Operations at the Pohang plant has been put on hold indefinitely. The energy policy had to be reviewed and priority lists were created based on risk assessments <sup>[27]</sup>. The perception change led to a ripple effect which ultimately had a marked impact on all stakeholders involved in the endeavor. As shown in the earlier section, once the public were made aware of the causal link between EGS and induced seismicity, they began to associate the technology with the damages incurred leading to its lowest position on the favorability chart <sup>[26]</sup>. Moreover, South Korean regulators have instigated high-level changes as indicated in the bow-tie analysis such as a focus on building materials as well as advanced geological surveys have been initiated

with standard operating protocols being laid out for each of them <sup>[27-28]</sup>. Another significant change motivated by the incident is the development of disaster insurance industry with demand for the same surging following the government investigation mentioned earlier <sup>[28]</sup>. This shows that the public have become more cognizant of the induced seismicity risk following the Pohang earthquake.

#### 6. Conclusion

The Pohang earthquake is an insightful case study that acutely details the dangers of premature deployment of a relatively nascent technology. The demand for sustainable alternative fuel sources is at an all-time high and this pressure is often devolved to technocrats and policy makers. Owing to South Korea's focused Energy Policy, regulators are required to expedite all proceedings pertaining to the same and this led to the oversight, as described in the earlier sections, which ultimately caused the induced seismic activity. Such instances force a scenario where risk identification and analysis are done for a limited scope. Thus, overarching risk factors and long-term repercussions are not integrated in operational design. Induced seismicity is a major issue that is often overlooked during geothermal extraction as past induced events often fell under the microseismic category. The Pohang earthquake measured significantly higher on the Richter scale and forced all responsible parties to reevaluate EGS. This served as the motivation for regulatory changes outlined in section 5.4. As depicted in the bowtie analysis in section 4, due deliberation at the preliminary stages could very well mitigate the impact of induced seismicity with onus being on comprehensive geological survey and fault identification. Optimizing operational parameters such as injection fluid composition, frequency and flow rates could also serve to minimize the impact of EGS. The major technical intervention required to ensure safe operations during EGS operations in the future are dynamic simulations and modelling as evidenced by the Pohang incident and described in the Bow-tie analysis. A combination of these technical and regulatory interventions holds the key to safe deployment of EGS technology.

The Pohang earthquake is also an important study of how improper risk management can distort public perception of a viable and sound technology leading to its shelving. In addition to financial losses and legislative bottlenecks, the sudden pause to a piece of critical innovation could significantly hamper progress in the particular discipline and very well eliminate it from the arsenal of sustainable development. The risk posed by induced seismicity has overshad-owed the benefits of geothermal energy and resulted in the indefinite suspension of the EGS plant. The Pohang incident highlights the importance of thorough and detailed risk management practices to ensure safe operations as well as the necessity to remain in the good books of the public, a vital parameter for the adoption of alternative energy.

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To whom correspondence should be addressed: Anil Kumar Vinayak, Department of Chemical & Materials Engineering, University of Alberta, Canada; <u>e-mail</u>: <u>anilkum1@ualberta.ca</u>

# Article

Investigation of the Potentials of Heat Extraction from Abandoned Oil Wells for Electricity Generation

Nkemakolam Chinedu Izuwa\*, Vincent Ebuka Udigwe, Onyebuchi Ivan Nwanwe\*, Ndubuisi Uchechukwu Okereke

Department of Petroleum Engineering, Federal University of Technology, Owerri, Nigeria

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#### Abstract

In the era of high energy demand caused by rapid population growth, renewable energy is required in the energy mix for the sustainability of both human and industrial development. This paper investigates the potential of extracting heat energy from abandoned oil and gas wells for electricity production. Four wells located in the Niger Delta were considered. Each well has its properties and steam flow rates. The geothermal reservoir was modeled as a liquid-vapor-dominated system at in situ reservoir conditions. The fluid temperature in the reservoir was determined to be 170°C in the reservoir. Equations describing geothermal resource potentials, energy flow in a wellbore, and gross electrical power production using turbines were presented. A Single-flash turbine system was used because steam was produced in the reservoir. MATLAB software was used in the simulation of the heat transfer process. The results showed that the geothermal energy corresponding to the reservoir considered was  $2.45 \times 10^{12}$  Joules. The electricity production result showed that there was a reduction in gross electrical power output due to an increase in sink temperature, while the relationship between gross electrical power output and separator temperature was such that there was an initial increase in gross electrical power output due to an increase in separator pressure until an optimum point was reached wherein further increase in separator pressure progressively decreased the value of the gross electrical power output. The optimum separator temperatures for well 1, well 2, well 3, and well 4 were 120°C, 120°C, 110°C, and 105°C respectively at a sink temperature of 45°C. Because of the high reservoir pressure used, the separator pressure was higher than the actual reservoir pressure. Proper understanding of heat flow which enables accurate estimation of heat production is necessary to maximize heat extraction from the reservoir. The simulation conducted reveals that geothermal resources holds great potentials for electrical energy generation and should be put to developmental use in Nigeria as a means to increase the total power generation and increase the portion of renewables in Nigeria energy value chain.

**Keywords**: Geothermal energy Extraction; Turbine; Abandoned oil and gas wells; Fluid dominated; Heat transfer; Electric power generation; Renewable energy.

#### 1. Introduction

Renewable energy resources have demonstrated great potentials as the most efficient and effective solutions to global environmental problems and with promises of long-term sustainable development. Diversified and sustainable energy resources are wholly indispensable in the fight against global climate change and looming world environmental issues <sup>[1]</sup> and global energy demand. The reduction in fossil fuel usage and increased attention to renewable and alternative energies such as solar, wind, geothermal energies are considered as a transition to a global green energy future (ultra-low or no carbon footprints). Geothermal energy can be regarded as the most promising sustainable energy given its abundant storage and reliable sources <sup>[1]</sup>.

Geothermal energy is renewable energy found beneath the earth's surface. It is a source of thermal energy whose generation and storage are within the earth's crust. The geothermal

energy stored in the earth's crust is inexhaustible considering human standards and needs. It has a much less expensive production capability and much pollutant-free during its production or extraction. Furthermore, another peculiar advantage of geothermal energy extraction is that it is not affected by weather conditions like solar and wind.

Geothermal energy can also be used in district heating and agriculture. A geothermal system consists of a reservoir, a heat source, and a carrier fluid that transfers the heat <sup>[2]</sup>. Thus a geothermal system can be grouped into three main systems namely hydrothermal, geopressured and hot dry rock systems depending on the heat transfer medium. Hydrothermal systems which are dominated by vapor and hot water are further classified as conventional geothermal resources while hot dry rocks are classified as non-conventional resources <sup>[3]</sup>. The unconventional resources have been shown to have higher potentials for geothermal development because they exist at very high temperatures and heat storage. The heat storage of hot dry rocks system is almost 168 times that of hydrothermal systems <sup>[2,4]</sup>.

The development of a geothermal resource requires the presence of a circulation fluid necessary to extract the heat from the reservoir to the surface. This involves a non-isothermal flow because of heat exchange <sup>[2]</sup>. Subsurface heat transfer can occur in conductive and convective heat transfer modes. Besides these, other prevalent uncertainties affect the distribution and transfer of heat. These factors could be reservoir-resident factors such as reservoir heterogeneities and geological structures and factors from the well layout and designs such as doublet conditions <sup>[2]</sup>. An accurate understanding of heat flow which enables proper estimation of heat production is necessary to maximize heat production from the subsurface <sup>[5]</sup>.

The electricity producible from geothermal resources depends fundamentally on the nature of underground resource conditions. Although geological assessment of in-situ thermal resources is available but the more reliable resource assessments are guaranteed after test wells are drilled to the subsurface. Electricity is produced from geothermal fluids using suitable Rankine cycle power plants. Three types of power plants are used for electricity generation from geothermal energy; these are the dry steam plants, the flash cycle plants, and the binary cycle plants <sup>[6]</sup>. The dry steam geothermal power plants are used for electricity generation when the geothermal resource produces hot dry steam directly from the well. The steam comes from the well and is first passed through separators that remove sands and rock particles before it is sent to the turbine. Flash steam plants are utilized on occasions where the geothermal resource produces high-temperature hot water or a combination of steam and hot water. The fluid from the well is delivered to a flash tank where a portion of the water flashes to steam and is directed to the turbine. Two-stage flash tanks could be utilized depending on the temperature of the fluids. They are the most common types of geothermal power plants due to the lack of naturally occurring high-quality steam. But just like the former, his type of plant cannot be applied to low-temperature resources <sup>[6]</sup>. Binary geothermal power plants use a second fluid in a closed cycle to operate the turbine rather than geothermal steam. Binary power plants use fluids with low boiling points such as butane or pentane in a secondary loop. The water flowing from the well through a heat exchange transfers its heat to this fluid which causes vaporization of these fluids due to their low boiling point. It is then passed through a turbine and used for electricity generation <sup>[7-8]</sup>.

There is evidence that supports the high initial cost of geothermal reservoirs when new fields are to be developed especially for geothermal resource extraction. This cost is mostly related to exploration and reservoir mappings present high upfront risk which may limit investment opportunities in geothermal resource extractions <sup>[9-10]</sup>. Because of this, suggestions have been made on the usage of abandoned oil and gas wells for geothermal resource extraction. These wells can be retrofitted for geothermal utilization. These wells were abandoned because petroleum recoveries from them were far below the marginal economic index. Although abandoned oil and gas wells can be used for geothermal extraction after being retrofitted with insulated production tubing, more attention is given to the usage of gas wells for this purpose. By retrofitting already existing wells for geothermal usage, initial investment costs from drilling and partially from completions would be obviated. Studies showed that abandoned oil and gas wells were beneficial to geothermal energy extraction <sup>[11-13]</sup>.

### 2. Geothermal energy

Geothermal energy represents the thermal energy that comes from the earth due to the temperature of the hot reservoir rock. This heat comes from the core of the earth and is transferred from the core largely through conduction through solid rock, and then through convection in places with fluid contact (e.g. water, magma, salt diaper) <sup>[14]</sup>. There is enormous energy present in the earth's crust that can be harnessed for future energy needs. The overall thermal energy in this earth is in the range of 12.6 x  $10^{12}$  EJ and 5.4 x  $10^{9}$  EJ in the range of crust up to a depth of 50 km <sup>[15]</sup>.

There has been an existence of geothermal energy residents in Nigeria's geographical location. Most of the geothermal resources in Nigeria come as warm/hot springs, seepages located mainly within the sedimentary basin of Benue Trough. The geothermal resources in Nigeria include the Ikogosi warm spring with a temperature of 37°C [16]. Rafin Reewa, is another warm spring in Jos Plateau with a temperature of 42°C and runs from. There have been shreds of evidence suggesting the existence of geothermal resources in the Anambra basin, Sokoto basin, Chad basin, Bida basin.

### 3. Methodology

This section presents the methods for the study. This comprises the development of models for geothermal systems and the simulation of the model in computer software.

### 3.1. Development of the models

This comprises the development of equations that governs the system and all processes involved. For this work, the equation development shall comprise three distinct parts, they are

- 1. Estimation of geothermal heat resource
- 2. Modeling the heat flow rate in the extraction wells
- 3. Electrical power calculation using turbines

### 3.1.1. Estimation of geothermal heat resource

The type of geothermal reserve determines the equation for the estimation of its heat capacity. The geothermal reservoir may contain dry steam, wet steam, dry rock as its source of heat. These have their peculiar models used in the estimation of the resident heat at reservoir conditions.

The heat transfer medium used in this research work is wet steam which comprises steam and hot water. The determination of the heat potential is therefore done for hot water and steam. The total heat in the reservoir is the sum of the heat of the two phases co-existing together in the reservoir.

The thermal energy from hot water in a geothermal reservoir is given below as  $E_{iw} = \phi_i \rho_w C_w V_i (T_i - T_o)$  (1)

where  $C_{wi}$  is the volumetric heat capacity of the hot water for ith volume;  $\rho_{fi}$  is the density of the hot water for the ith volume.

Similarly, if the fluid in the reservoir contains hot water and steam, then a two-phase system co-exist and the overall fluids thermal energy is given as

$$\begin{split} E_{if} &= \varphi_i \rho_w C_w V_i (T_i - T_o) + \ \varphi_i \rho_s C_s V_i (T_i - T_o) \end{split} (2) \\ \text{where } C_{si} \text{ is the volumetric heat capacity of the steam for ith volume; } \rho_{si} \text{ is the density of the steam for the ith volume.} \end{split}$$

There are some systems where the heat sources are a combination of rock and fluid energy. In this system, thermal energy is a contribution of the heat from the rock and the fluid. The thermal energy for this reservoir system is estimated below  $F_{1} = F_{1} + F_{1}c$ (3)

$$E_{i} = E_{ir} + E_{if}$$
This can further be expanded to become
$$E_{i} = (1 - \phi_{i})\rho_{ri}C_{ri}V_{i}(T_{i} - T_{o}) + \phi_{i}\rho_{w}C_{w}V_{i}(T_{i} - T_{o})$$
(4)

To calculate the energy in the whole reservoir, integration of the entire reservoir is done. If the reservoir is composed of n volume components, then the total thermal energy in the reservoir is given as

 $E_R = \int_i^n E_i dv$ Note that

 $V = V_i + V_k + - - - V_n$ 

(5)

(8)

(10)

(6)Not all the thermal energy in the reservoir is recoverable. The amount of heat recoverable is a function of the extraction technology and method of extraction applied. The recovery factor is the ratio of the total thermal energy recoverable from the reservoir to the total thermal energy in place in the reservoir at initial conditions. The recovery factor is given as R  $R = \frac{E_s}{E_R}$ (7)

 $E_s$  is the thermal resource extractable at the current technological and economic condition;  $E_R$ is the total thermal resource in the reservoir.

 $E_{\rm s} = E_{\rm R} n$ 

### 3.1.2. Modeling the heat flow in the extraction wells

The energy exchange equations which fundamentally comprises the thermal variations and heat transfers along with the extraction well for wet-steam dominated geothermal reservoirs is given in this section

The equation for energy in the extraction fluid well is given in equation 9  $\frac{dT_{ex}}{dt} + \frac{d(VT_{ex})}{dz} = -S_{exin}$ (9) dt

where Tex fluid temperature in extraction well; V fluid flowrate; z vertical distance from earth surface; t time interval; Sexin heat exchange rate between extraction well.

 $S_{inex} = \frac{k_l(T_{in} - T_{ex})}{k_l(T_{in} - T_{ex})}$  $\rho A_{ex}C_{p}$ 

where  $\rho$  density of the fluid; A<sub>ex</sub> Flow area of the extraction well; C<sub>p</sub> Specific heat of the fluid in the extraction well; k heat conductivity coefficient for unit length. (11)

 $A_{ex} = \pi r_{ex}^2$ 

where rex the radius of the reservoir.

# 3.1.3. Electrical power calculation using turbines

This section deals with the calculations involved in the conversion of the thermal energy in the geo-fluid to electrical power using turbine systems. Two major types of turbines are useful with a geo-fluid in production of electricity: they are the flash turbine system and the binary turbine system

# i.) Installed power

The Installed power gotten from a geothermal plant due to thermal energy from a geothermal resource is given as

 $P_i = \frac{E_R C_e}{f_l t}$ 

(12)

where  $E_R$  is recoverable heat, KJ;  $P_i$  is installed power, MWe;  $C_e$  is conversion efficiency of plant; fi is load factor; t is commercial lifespan of the plant, Msec.

# ii.) Flash system turbine calculation

Because the geothermal resource utilized in this study is fluid dominated saturated steam system, a flash turbine system would be the most appropriate turbine type for use in the generation of electricity using the geo-fluid

Figure 1 illustrates the temperature-entropy diagram of a single flash turbine system.



Figure 1. Temperature-Entropy diagram for single flash turbine system for geothermal heat conversion to electricity

The single flash turbine system comprises several processes; these include the flashing process, the separation process, the expansion process, and the condensation process. The block diagram in Figure 2 illustrates the sequences in the single flash turbine process.



Figure 2. Block diagram of single flash process

The electrical power output of the flash turbine system can be given from Figure 2 as  $w_e = x_2 m_{total} (h_4 - h_{5s}) \eta_t \eta_g$  (13) where  $m_{total}$  is total flowrate of the mixture of vapor and hot water at point 2;  $\eta_g$  is generator efficiency;  $\eta_t$  is isentropic efficiency of the turbine;  $x_2$  is ratio of steam to liquid.  $x_2 = \frac{h_2 - h_3}{h_4 - h_3}$  (14)

### 3.2. Case study

The case study used in this work is four oil and gas wells in field Asa-2 in the Niger Delta. The wells are wells AX-2, AX-3, AX-5 and AX-7. These are abandoned oil and gas wells that have been converted to geothermal wells and used for heat extraction. This was chosen to reduce the cost of exploration and drilling that would be associated with developing a new geothermal well that was not previously drilled. Using an abandoned oil and gas well limits the economic expenses in terms of exploration, drilling, and well completion. Except that the well has to be retrofitted to accommodate the high-temperature fluid recovery. An insulated

production liner will be used. By this, the total cost of producing a unit of electricity from the geothermal reservoir would be greatly decreased.

The reservoir data and the fluid data at the wellhead and surface equipment respectively as well as the parameters for the turbine system used are given below in tables 1 and 2. The properties of each well are given in table 3

|--|

Parameter	Units
Fluid Reservoir temperature, °C	170 ℃ (338 °F)
Reservoir Pressure, psia	4300
Fluid temp @wellhead, °C	197.2
Fluid pressure @ wellhead, bar	227.91 psia
Porosity	0.25
Permeability, mD	600
Area of Reservoir, acres	100
Reservoir thickness, ft	200
Density of steam in reservoir, kg/m <sup>3</sup>	11.57
Density of hot water, kg/m3	960
Heat capacity of hot water, kJ/kg.K	4.2
Heat capacity of steam, kJ/kg.K	3.408
Environmental temperature, °C	30

Points	Enthalpy, kJ/kg	Entropy, kJ/kg.K
1	839.9	2.203
2	839.9	2.203
3	482.5	1.463
4	269.9	7.1842
5	2356.7	-
5s	2271.16	7.1842
6	-	0.6387
7	-	8.1648

Table 2. Fluid parameters

Table 3. Properties of each well

Parameter	Well Values					
ralameter	Well AX-2	Well AX-3	Well AX-5	Well AX-7		
wellhead pressure, psig	200	200	200	200		
wellhead temperature, °C	197.2	206	177.7	171.4		
skin	3	3.5	4	3.7		
tubing size, in	3.958	3.958	3.958	3.958		
well radius, ft	0.56	0.56	0.56	0.56		
geothermal gradient, °F/100ft	4.17	4.39	3.68	3.53		
well depth, ft	9890	10100	11,200	11,800		

# 3.3. Construction of the geothermal field production

GAP was used to construct the field geothermal wells resource production. There are a total of four wells that were previously used for oil and gas production. The wells were converted and used for geothermal heat extraction. The flow diagram of the extraction wells is given in the Figure 3.

All the fluids are extracted from one reservoir. Each of the fluids flow to the wellheads. All the wellhead fluids are channeled to the production manifold where they are then routed to the separator. Flash takes place between the wellheads and the separator.



Figure 3: Well and surface lines arrangement

# 3.4. Simulation approach

The simulation was done by building models in MATLAB. This is done by writing Matlab scripts files that do essentially three basic things.

- a. Estimates the recoverable heat in the reservoir given the geothermal system type
- b. Estimates the wellbore heat loss from the reservoir to the surface given the wellbore and surface equipment used



3.5. Well model



Figure 5. Schematic diagram of the modeled geothermal system

the well was completed with 9 5/8 in casing.

After the wells were abandoned, before plugging the wells, they were converted to geothermal wells. This was done through retrofitting and the addition of insulators at the casing to prevent heat losses from the reservoir to the surface. The insulation was achieved by sealing off the bottom of the casing with non-conductive materials. A schematic diagram of the model is given in Figure 5. Figure 6 shows the vertical section of the geothermal well. It can be observed that



Figure 6. Vertical profile of the well AX-2

# 4. Results and discussions

### 4.1. Geothermal resource potential



The Geothermal heat resource potential from the simulation performed is 2.45 x  $10^{12}$  Joules. This was determined using equation 4 and 5 .But not all of this heat can be extracted. The heat potential as a function of reservoir temperature is given in the Figure 7. From Figure 7, it can be observed that the geothermal heat potential of a geothermal reservoir increases with an increase in reservoir temperature. As the wet steam is produced, the pressure of the reservoir begins to decrease.

Figure 7. Thermal potential as a function of reservoir temperature

# 4.2. Specific gross electrical energy per molar flow rate of steam

The specific gross electrical power from the turbine from each of the four wells corresponding to their various wellhead fluid temperatures is given in this section. The specific gross electrical power is the gross electrical power per mass flowrate of steam from each wellhead for the four wells considered. The flash turbine system simulated produced gross electrical power per mass flowrate of steam for various separator temperatures and condenser (sink) temperatures.

# a) Well 1: Wellhead fluid temperature of 167°C

The specific gross electrical power is given in the Table 4 for well 1

Separator tem-	S	pecific gross electrical	power, kW/(kg/s)	
perature, °C	Sink Temp= 45°C	Sink Temp= 50°C	Sink Temp= 55°C	Sink Temp= 60°C
95	44.39	39.76	35.17	30.63
100	46.13	41.73	37.38	33.07
105	47.36	43.20	39.07	35.00
110	48.27	44.34	40.43	36.58
115	48.67	44.96	41.29	37.66
120	48.74	45.25	41.81	38.39
125	48.32	45.06	41.85	38.66
130	47.58	44.55	41.55	38.59
135	46.38	43.57	40.80	37.61
140	44.84	42.27	39.71	37.18
145	42.88	40.52	38.18	35.88
150	40.58	38.45	36.33	34.24

Table 4.	Well	1 Speci	fic gross	s electrical	power	output
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The specific gross electrical power output for well 1 is given in Table 4. This is done for several separators and sinks temperatures. The shaded areas in Table 4 correspond to the maximum power output for the conditions specified. For sink temperatures of 45°C and 50°C, the maximum electrical power was 48.74 kJ/kg/s and 45.25 kJ/kg/s at a separator temperature of 120°C. But for sink temperatures of 55°C and 60°C, the maximum electrical power was 41.85 kJ/kg/s and 38.66kJ/kg/s at separator temperature 125°C

It can be observed from Table 4 that the specific gross electrical power produced increased with an increase in separator temperature for all condensers (sink temperatures) to a maximum point wherein it starts to decline. This point is the optimum operating condition of the turbine and must be determined for all turbine systems using geofluids.

#### b) Well 2: Wellhead fluid temperature of 165.5°C

The specific gross electrical output per mass flowrate of steam for well 2 is given in the Table 5

Separator tem-		Specific gross electri	ical power, kW/(kg/s	)
perature, °C	Sink Temp= 45°C	Sink Temp= 50°C	Sink Temp= 55°C	Sink Temp= 60°C
110	53.28	48.93	44.64	40.39
115	54.02	49.90	45.83	41.81
120	54.43	50.54	46.70	42.89
125	54.35	50.69	47.07	43.49
130	53.95	50.51	47.12	43.76
135	53.08	49.86	46.69	43.54
140	51.88	48.89	45.93	43.01
145	50.23	47.47	44.73	42.03
150	48.27	45.72	43.21	40.72
155	45.85	43.54	41.24	38.97
160	43.13	41.02	38.96	36.90
165	39.96	38.09	36.24	34.41

Table 5. Well 2 electrical power output

This is done for several separators and sink temperatures. The shaded areas in the table correspond to the maximum power output for the conditions specified. For sink temperatures of 45°C and 50°C, the maximum electrical powers were 54.43 kJ/kg/s and 50.54 kJ/kg/s at a separator temperature of 120°C. But for sink temperatures of 55°C and 60°C, the maximum electrical power was 47.12kJ/kg/s and 43.76 kJ/kg/s at separator temperature 125°C.

### Well 3: Wellhead fluid temperature of 164.7°C

The specific gross electrical power for well 3 is given in Table 6 for several separator and condenser (sink) temperatures.

Separator tem-	Specific gross electrical power, kW/(kg/s)						
perature, °C	Sink Temp= 45°C	Sink Temp= 50°C	Sink Temp= 55°C	Sink Temp= 60°C			
95	35.67	31.95	28.26	24.62			
100	36.61	33.12	29.52	26.24			
105	37.08	33.82	30.59	27.40			
110	37.20	34.17	31.16	28.19			
115	36.85	34.04	31.26	28.52			
120	36.15	33.57	31.02	28.49			
125	35.01	32.64	30.31	28.01			
130	33.52	31.38	29.26	27.19			
135	31.59	29.67	27.78	25.92			
140	29.32	27.64	25.97	24.31			
145	26.65	25.18	23.73	22.30			
150	23.63	22.38	21.15	19.93			

Table 6. Well 3 electrical power output

This is done for several separators and sink temperatures. The shaded areas in the Table correspond to the maximum power output for the conditions specified. For sink temperatures of 45°C and 50°C, the maximum electrical powers were 37.20 kJ/kg/s and 34.17 kJ/kg/s at a separator temperature of 110°C. But for sink temperature of 55°C and 60°C, the maximum electrical power was 31.26 kJ/kg/s and 28.52 kJ/kg/s at separator temperature 115°C **c) Well 4: Wellhead fluid temperature of 161.4°C** 

	Specific Gross Electrical Power, kW/(kg/s)					
Separator tem-		Sink Temp=	Sink Temp=	Sink Temp=		
perature, °C	Sink Temp= 45°C	50°C	55°C	60°C		
90	31.78	28.11	24.48	20.88		
95	32.89	29.46	26.06	22.70		
100	33.57	30.38	27.20	24.07		
105	33.80	30.82	27.88	24.97		
110	33.67	30.93	28.20	25.52		
115	33.08	30.56	28.06	25.60		
120	32.13	29.84	27.57	25.32		
125	30.75	28.67	26.63	24.61		
130	29.03	27.18	25.34	23.54		
135	26.87	25.24	23.64	22.04		
140	24.37	22.97	21.58	20.21		
145	21.46	20.28	19.11	17.95		

Table 7: Well 4 electrical power output

The electrical power output for well 4 is given in Table 7. This is done for several separators and sink temperatures. The shaded areas in Table 7 correspond to the maximum power output for the conditions specified. For sink temperature of 45°C, the maximum specific gross electrical power output was 33.80 kJ/kg/s at a separator temperature of 105°C; for sink temperature of 60°C and 50°C, the maximum specific electrical power output is 30.93 kJ/kg/s and 28.2 kJ/kg/s respectively all at separator temperature of 110°C. Similarly, for sink temperature of 60°C, the maximum specific gross electrical power output was 25.60 kJ/kg/s at a separator temperature, the electrical power out decreases as the condenser temperature increases (sink temperature).

#### 4.3. Actual gross electrical power for steam flowrates

The gross electrical power for steam flowrates according to each well's capacity is given in Figure 8 for sink (condenser) temperature of  $45^{\circ}$ C. As can be observed from the plot, the gross electrical power output produced is highest for well 2( 2.99 MW at 105°F) and lowest for well 4 ( 2.20 MW at  $110 ^{\circ}\text{F}$ ).

The gross electrical power for steam flowrates according to each well's capacity is given in Figure 9 for sink (condenser) temperature of 50°C. As can be observed from the plot, the gross electrical power output produced is highest for well 2 (2.79 MW at  $110^{\circ}$ F) and lowest for well 4 (2.01 MW at 115°F).





eral wells at 50°C sink temperature

The gross electrical power for steam flowrates according to each well's capacity is given in Figure 10 for sink (condenser) temperature of 55°C. As can be observed from the plot, the gross electrical power output produced is highest for well 2 (2.79 MW at  $110^{\circ}$ F) and lowest for well 4 (2.01 MW at 115°F).

The gross electrical power for steam flowrates according to each well's capacity is given in Figure 11 for sink (condenser) temperature of 60°C. As can be observed from the plot, the gross electrical power output produced is highest for well 2 (2.79 MW at 110 °F) and lowest for well 4 (2.01 MW at 115°F).



Figure 10. Gross electrical Power output for several wells at 55°C sink temperature



Figure 11. Gross electrical Power output for several wells at 60°C sink temperature

### 4.4. Discussion of result

Comprehensive simulation of the geothermal potential in the development of geothermal reservoirs has been carried out. From section 3.1, the geothermal heat resource potential was given. This was calculated from equation (2) in chapter three because the source of heat for this work is steam and hot water at in-situ reservoir conditions. Because of this vapor-liquiddominated geothermal system, a single flash turbine was used in electricity production. For

each well, the wellhead pressure and temperature were given and these were used to determine the wellhead temperature corresponding to the saturated fluid from steam tables.

The electricity production was calculated in two stages. The first was done per unit mass of steam flow rate to get the specific gross electrical power output while the second gave the total electrical power produced as a result of each well's steam flowrate produced in the separator. The electrical power output was crucially determined by the temperature of the separator and the sink (condenser) and the flow rate of the steam flashed and separated in the separator.

Well 2 because of its high temperature gave the highest gross electrical power for all cases of sink pressures and separator pressure.

From the sensitivity analyses conducted using sink pressures; it was observed that the gross electrical power output decreased with an increase in sink temperatures. But for the case of separator pressure, the gross electrical power output initially increased with an increase in separator pressure until a maximum value is reached wherein the gross electrical power output begins to decrease with an increase in separator temperature.

#### 5. Conclusions

Modeling of geothermal reservoir potential using depleted oil and gas wells has been conducted. The abandoned oil/gas well was necessary to offset investment costs in drilling and completion of new wells for geothermal resource exploitation thereby improving the economic feasibility of the project and the affordability of the electrical resource produced in the process.

The process was done to quantify the geothermal resource potential and possible electrical energy produced from the flowrate and pressure-temperature conditions of the wellhead saturated geofluids produced. A single flash turbine system was used and the electrical power produced was given unit of a mass flow rate of steam (kJ/kg/s) and in actual electrical power as a result of each well's steam flowrate produced (in MW). The geothermal resource was estimated as  $2.45 \times 10^{12}$  Joules. The optimum separator temperature obtained for the wells are  $120^{\circ}$ C,  $120^{\circ}$ C,  $110^{\circ}$ C, and  $105^{\circ}$ C for well 1, well 2, well 3, and well 4 respectively. The electrical power produced decreased with increasing sink temperature. Thus  $45^{\circ}$ C was the best sink temperature because it led to the highest electric power generation.

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To whom correspondence should be addressed: Dr. Onyebuchi Ivan Nwanwe, Department of Petroleum Engineering, Federal University of Technology, Owerri, Nigeria, Nigeria; <u>e-mail</u>: <u>onyebuchi.nwanwe@futo.edu.ng</u>

# Article

# **Open Access**

Comparative Analysis of Water Quality Assessment in the Basement Complex terrain of Central Nigeria: A Case Study of Ground and Surface Water Relations in the Agbadu - Bunu Community and it's Environ in Kabba/Bunu Local Government Area of Kogi State

Seyi Mepaiyeda<sup>1, 2</sup>, Christopher Baiyegunhi<sup>3</sup>, Omonayin Obafemi<sup>2</sup>, and Siyaka Abdulsalam<sup>2</sup>

- <sup>1</sup> Department of Geology, of Faculty Science and Agriculture, University of Fort Hare, Private Bag X1314, Alice 5700, Eastern Cape Province, South Africa
- <sup>2</sup> Department of Mineral Resources Engineering, School of Engineering, Kogi State Polytechnics, PMB 101, Lokoja, Kogi State, Nigeria
- <sup>3</sup> Department of Geology and Mining, School of Physical and Mineral Sciences, University of Limpopo, Private Bag X1106, Sovenga 0727, Limpopo Province, South Africa

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#### Abstract

Analysis of water resources around Agbadu - Bunu Community in Kogi State, Nigeria was carried out to assess the water quality by determining the concentrations of cations, anions, heavy metals, trace elements, pH, alkalinity, total dissolved solids and microbial loads in water samples. Ten water samples (one rainwater, two pond waters, one stream water, four hand-dug wells water, one hand pump water and one motorized borehole water) were systematically collected in the study area and analyzed using the atomic absorption spectrophotometer. Microbiological analysis for total bacteria count (TBC), total coliform (TC) and other intestinal bacteria were assessed using standard plate count Agar, Macconkey Agar, Macconkey broth and Brilliant Green Bile Agar. Multiple bottle methods were employed in the enumeration of Coliform. The physiochemical results were compared with the standard values recommended by the World Health Organization (WHO) and it showed that all the cations in water samples are within the WHO recommended limits. The water samples at locations B, E, F, G, H, J, and I have high alkalinity and HCO<sup>3-</sup> content. The heavy metals (Pb<sup>2+</sup>, Ni<sup>2+</sup> and Cd<sup>2+</sup>) and trace elements (Fe and Cu) are dominantly high in the surface waters. The analysis of microbiological substances in the water samples revealed that most of the water in the area are not fit for human consumption due to the presence of indicator organisms higher than the WHO standard for drinking water. However, Samples obtained from locations G, I and J have TBC values of 40cfu/100mL, 10cfu/100mL and 05cfu/100mL, respectively. In addition, no E-Coli was recorded in these locations and they have the same TC values of 0cfu/100mL. The TBC and TC values fall within the WHO recommended limit, thus they are fit for human consumptions but needed to be treated due to their high MPN (Most Probable Number) index of <11/100 mL, <24/100 mL and <26/100 mL, respectively as against WHO standard of <10/100 mL. This study shows that the water resources in the study area are gradually been polluted and in the nearest future may not be good for drinking.

Keywords: Water; Rainfall; Heavy metals; Anions; Microbes; Nigeria.

### 1. Introduction

A large number of communities in Nigeria, especially those in the Eastern highland, Western highland and central parts depend largely on groundwater supply from the Basement Complex rocks through boreholes. The development of groundwater resources in these parts of Nigeria are carried out under severe budgetary constraints, with little opportunity for fundamental hydrogeological studies. Hard rock masses such as the basement-rock masses covering about two-third of the land surface of Nigeria in their under -formed state possess little or no primary inter-granular porosity or permeability and the hydro-geological properties are thus mainly

determined by secondary storability and transmissivity. It is not possible to find absolutely pure water in nature even rainwater, when it drops, just emitting from the clouds, may be considered pure, but as the drops fall down, certain gasses get dissolved in it and make the rainwater impure <sup>[1]</sup>. Depending on the sources of the water, surface and groundwater may differ greatly in terms of purity and suitability for the purposes for which they are required <sup>[2]</sup>. Groundwater is the water, which is stored by nature, underground in the water bearing formations of Earth's crust. This could be natural springs, well and boreholes, infiltration galleria and radial collector wells <sup>[2]</sup>. The inorganic materials for which maximum contaminant levels have been established are generally toxic in one manner or the other. Lead, mercury, arsenic, barium, cadmium and chromium are poisonous and can exhibits chronic or acute toxicity depending on the concentration <sup>[3-7]</sup>.

In the study area, groundwater occurrence is in the weathered basement or in the joint and fracture systems of the unweathered or partly weathered rocks. There are aquifers in the Basement Complex, hence, each site has to be treated as unique. The depths of aquifers are normally between 10 m and 60 m. Previous work by Omada *et al.* <sup>[5]</sup> on the physico-chemical characteristics of surface and ground water in central Nigeria shows that the concentration of cations and anions in the water resource of certain parts of central Nigeria conformed to the World Health Organization (WHO) for potable water. Omada *et al.* <sup>[5]</sup> collected fifty-five water samples for both cations and anions analysis using inductively coupled plasma-optical emission spectrometry (ICP-DES) and inductively coupled plasma-mass spectrometry (ICP-MS). They reported that that the area (central Nigeria) consist of two water types, namely, the earth-alkaline water and earth-alkaline waters. Furthermore, they documented that alkaline compounds with cations and anions concentrations are below WHO limit in some parts while other parts had their concentrations above WHO limits. As a result of the reported variations in the water types, in this study, we examine the quality of drinking water around Agbadu-Bunu in Kabba Local Government Area of Kogi State, central Nigeria (Figure 1).



Figure 1. Map of Nigeria showing the study area and sample locations

The area has a fast growing population and increasing level of diverse socio-economic activities. Factors responsible for water contamination and the type of contaminants in water to determine its deviance from the normal were also investigated. The study area lies between latitude 7.91786<sup>o</sup> E and longitude 6.26911<sup>o</sup> N. The community experiences distinct dry and wet seasons. There is a fairly high amount of rainfall in the wet season and water is readily available during the wet season but may become dried or dry up during the dry season. This is particularly so in areas underlain by crystalline rocks of the Basement Complex where many rivers are intermittent and the underlain lithology does not favour large accumulation of groundwater <sup>[4]</sup>.

### 2. Material and methods

A preliminary (reconnaissance) survey of the study area was carried out in order to study the various sources and kind of drinking water available in the area. This survey helped in the locations of various water sources and was marked out for random sample collection. Global positioning system (GPS) was used to measure the longitude and the latitude of the respective sample locations as well as the distance above sea level (Figure 1; Table 1). Two different types of containers were used for sample collection. Bottles for microbiological samples were first washed with detergent, rinsed with distilled water, dried and sterilized at 150°C for 2 hours, while other plastic bottles were washed with detergent, rinsed with distilled water before used for water sampling. In the process of sampling, care was taken that the container used in fetching the water (fetcher) was thoroughly washed to make sure that there is no contamination during sampling or level of contamination is minimized. For the rainwater sources, early morning water from the rain was collected into a sample bottle. Boreholes water samples (hand pump water and motorized borehole) were collected after the mouth of the tap was first swabbed/sterilized with cotton wool soaked with alcohol (ethanol). Some quantities of the water were first pumped out to create room for fresh water from the source and also help in flushing out bacteria or other possible contaminants along the lining of the pipe. The water samples taken from both surface (stream, rain, pound) and ground (hand dug wells/boreholes) water were collected at random and labeled accordingly After collection, the samples were protected from reacting with air by tightening the cork properly, packed into a small cooler and transported immediately to the laboratory for analysis. The samples were analysed using the atomic absorption spectrophotometer (AAS). The different methods used were in accordance with the hydrological project technical assistance (HPTA) method for standard analytical procedure for water. The concentrations of cations, anions, heavy metals and trace elements were determined. The pH of the samples was determined using a pH meter (Mode: HP 2211 PWORP meter). 10 mL of each of the sample was poured in to a sterile beaker and the anode of the pH meter was inserted into it, allowed to stay till for about 5 minutes before the readings were taken and recorded one after the other. The stainless sensor was rinsed with distilled water after each reading.

Sample ID			Coordinates		
ID	Label	Type of sample	Longitude	Latitude	
Α	RW	Rain Water	N 7º8′5.69″	E 6º2'5.65"	
В	PW1	Pond Water	N 7°55.6′55″	E 6º1'5.75"	
С	PW2	Pond Water	N 7°55.6′55″	E 6º1'5.75"	
D	SW	Stream Water	N 7º55.4′77″	E 6°16′32″	
E	HDW1	Hand Dug Well Water	N 7°55′25″	E 6º1.6'38"	
F	HDW2	Hand Dug Well Water	N 7°55′51″	E 6º16.1'60"	
G	HDW3	Hand Dug Well Water	N 7º55′55″	E 6º15.7′52″	
Н	HDW4	Hand Dug Well Water	N 7°55′087″	E 6º15.7′57″	
Ι	HPW	Hand Pump Water	N 7°55′27″	E 6°1.6′14″	
J	MPW	Motorized Pump Water	N 7°55′03″	E 6°1.6'13"	

Table 1. Sample points and their coordinates.

### 3. Results

The results of the total and average concentrations of cations, anions, heavy metal, trace elements, pH, alkalinity and total dissolved solids in the water resources (both surface and ground water) are presented in Tables 2 and 3. Table 4 shows the comparisons of the average concentrations of cations, anions, heavy metals, trace elements and pH, alkalinity and total dissolved solids of the water resources in the study area with the WHO standard for drinking water.

Table 2. Concentrations of cations, anions, heavy metals, trace elements, values of pH, alkalinity and<br/>total dissolved solids (TDS) in the water resources of the study areaParame-ABCDEFGHIJ

Parame-	A	B	C	D	E	F	G	Н	I	J
ter	(RW)	(PW1)	(PW2)	(SW)	(HDW1)	(HDW2)	(HDW3)	(HDW4)	(HPW)	(MPW)
(mg/L)										
Ca <sup>2+</sup>	4.38	4.22	4.21	3.64	4.12	4.101	2.90	2.64	2.11	1.48
Mg <sup>2+</sup>	2.64	2.50	2.49	2.103	1.94	1.90	1.81	1.92	2.53	1.21
K <sup>+</sup>	3.51	4.11	3.04	3.61	2.11	2.38	2.04	2.87	3.31	3.73
Na <sup>+</sup>	0.13	0.782	0.63	0.67	0.37	0.13	0.48	0.38	3.41	4.76
N0⁻₃	4.53	0.27	0.019	15.943	0.019	0.016	0.024	0.016	0.142	0.39
HC0⁻₃	19.76	66.15	48.97	38.67	103.95	126.30	59.30	64.43	251.72	97.08
CL <sup>-</sup>	29.78	6.62	8.51	17.02	16.54	17.02	25.53	25.52	19.85	23.16
S0⁻₄	5.32	3.801	3.803	0.25	0.38	0.36	0.38	0.39	0.25	0.66
Pb <sup>2+</sup>	0.031	0.071	0.07	0.30	0.044	0.043	0.046	0.05	0.034	0.031
Zn <sup>2+</sup>	0.16	0.22	0.23	0.13	0.12	0.104	0.101	0.102	0.12	0.002
Ni <sup>2+</sup>	0.071	0.06	0.051	0.35	0.03	0.03	0.039	0.03	0.045	0.02
Cd <sup>2+</sup>	0.004	0.075	0.073	0.062	0.04	0.05	0.041	0.06	0.014	0.011
Fe	0.74	0.241	0.25	0.42	0.13	0.12	0.14	0.12	0.33	0.19
Mn	0.01	BDL	BDL	BDL	0.00	0.00	0.00	0.00	0.00	0.00
Cu	2.01	3.46	3.61	2.88	2.19	2.18	2.18	2.19	2.41	0.70
PH	6.95	7.18	6.96	7.06	7.22	7.01	6.82	6.76	7.21	7.66
Alkalinity	43.33	133.33	100.00	80.00	206.67	250.00	120.00	130.00	493.33	193.33
TDS	0.00	20.00	20.00	20.00	380.00	20.00	20.00	20.00	360.00	20.00

*N.B.* Sample A = Rainwater (RW); Samples B and C = pond water (PW1 and PW2); Sample D = Stream water (SW); Samples E, F, G, and H= Hand dug well water (HDW1 – 4); Samples I and J = Boreholes (Hand pump well water (HPW) and motorized borehole water (MPW))

Table 3. Average concentrations of cations, anions, heavy metals, trace elements, values of pH, alkalinity and total dissolved solids (TDS) in the surface water resources of the study area

Parameters	Average concentrations	Average concentrations		
	in surface water (mg/L)	in ground water (mg/L)		
Ca <sup>2+</sup>	4.113	2.892		
Mg <sup>2+</sup>	2.433	1.885		
K+	3.568	2.743		
Na <sup>+</sup>	0.553	1.588		
N0₃ <sup>-</sup>	5.188	0.101		
HC03 <sup>-</sup>	43.388	116.13		
Cl-	15.483	21.268		
S04 <sup>2-</sup>	3.293	0.403		
Pb <sup>2+</sup>	0.118	0.041		
Zn <sup>2+</sup>	0.054	0.036		
Ni <sup>2+</sup>	0.185	0.92		
Cd <sup>2+</sup>	0.133	0.398		
Fe	0.213	0.172		
Mn	0.003	0.00		
Cu	2.99	1.975		
P <sup>H</sup>	7.04	7.113		
Alkalinity	89.165	232.22		
TDS	15.00	136.67		

Table 4. Comparison	of average concentrations of cations, anions, heavy metals, trace ele-
ments, values of pH,	alkalinity and total dissolved solids (TDS) in the water resources of the
study area with WHO	standards

Cations	WHO,2011 guideline maximum value mg/L	Guideline value mg/L	Maximum ad- missible con- centration mg/L	Average value of study area mg/L (This study)
Calcium (Ca <sup>2+</sup> )	-	100.00	-	3.503
Sodium (Na <sup>+</sup> )	200.00	20.000	150.000	1.071
Potassium (K <sup>+</sup> )	-	10.000	12.000	3.156
Magnesium (Mg <sup>2+</sup> )	-	30.000	50.000	2.159
Chloride ion (Cl <sup>-</sup> )	250	-	-	18.376
Sulphate ion $(S0_4^{2-})$	250	-	-	1.848
Nitrate ion $(NO_3^-)$	50	-	-	2.645
Hydrogen trioxocarbonate ion $(HC0_3^-)$	50	-	-	80.209
Lead (Pb <sup>2+</sup> )	0.010	-	0.005	0.080
Cadmium (Cd <sup>2+</sup> )	0.003	-	0.005	0.045
Zinc (Zn <sup>2+</sup> )	3.000	0.100	-	0.139
Nickel (Ni <sup>2+</sup> )	0.020	-	-	0.266
Iron (Fe)	0.300	0.050	0.200	0.193
Manganese (Mn)	0.050	-	-	0.002
Copper (Cu)	0.05	-	-	2.483
Ph	8.500	6.5-8.5	6.5-8.5	7.082
Alkalinity	100.000	-	-	174.999
TDS	500.000	-	-	88.000

Total Bacterial Count (TBC) value of the water samples analyzed has the mean value of 199.75 ×10<sup>3</sup> cfu/100 mL and 50 × 10<sup>3</sup> cfu/100 mL for surface and ground water of the area respectively. The highest values ranging from  $2.35 \times 10^2$  cfu/100 mL to  $1.38 \times 10^2$  cfu/100 mL for surface and ground water respectively against the 100 cfu/mL for WHO standard for potable water. Total Coliform Count of the water samples has mean values of  $2.0 \times 10^1$  cfu/100 mL and  $0.717 \times 10^1$  cfu/100 mL for surface and ground water respectively. The highest values ranging from  $0.23 \times 10^2$  cfu/100 mL to  $0.30 \times 10^2$  cfu/100 ml for surface and ground water, respectively against zero (0) cfu/100 mL for WHO standard for potable water (Table 5). E. Coli Count for the water samples analyzed is absent in most of the samples and only present in very minute amount in some of the samples. Though no particular amount was recorded, this mostly conformed with the zero (0) cfu/100 mL for WHO standard for potable water.

Table 5. Microbiological analysis of water samples in the study area

Microbiological/Sam-	A	В	С	D	E	F	G	Н	I	J
ples factors	RW	PW1	PW2	SW	HDW1	HDW2	HDW3	HDW4	HPW	MBW
Protozoan (count/mL)	0	12	0	0	0	0	0	0	0	0
Total plate count(cfu/mL)	5	110	129	235	50	138	40	57	10	05
Total coliforms (cfu/100mL)	10	30	17	23	30	03	0	10	0	0
Salmonella typhi	Absent	Absent	Absent	Absent	Absent	Absent	Absent	Absent	Absent	Absent
Shigella SP	Present	Absent	Absent	Absent	Absent	Present	Absent	Present	Absent	Absent
E. coli	Absent	Present	Present	Present	Present	Present	Absent	Present	Absent	Absent
Streptococcus faecalis	Absent	Absent	Absent	Absent	Absent	Absent	Absent	Absent	Absent	Absent
MPN Index<10/100 mL	2400	2400	94	8	2400	920	11	2	2400	26
	Accept	Not ac- cepta- ble	Accept	Not ac- cepta- ble	Accept	accept				

# 4. Discussion

The quality of water is most often a function of the mineralogical and geochemical characteristics of the rocks underlying the area <sup>[8]</sup>. Most minerals in rocks are soluble under appropriate geochemical condition. The quality of groundwater therefore, in some parts of the country, particularly shallow ground changed as a result of human activities. Groundwater is less susceptible to bacterial pollution than surface water because the soil and rocks through which groundwater flows screens out most bacteria <sup>[9]</sup>. Bacteria however occasionally find their way into groundwater sometimes in high concentrations. But, freedom from bacteria's pollution alone does not mean that the water is fit for drinking. Many unseen dissolved minerals and organic constituents are present in groundwater in various concentrations. Most of which are harmless or even beneficial, while others are harmful and a few may be highly toxic. Water typically is not considered desirable for drinking if the quantity of dissolved minerals exceeds 1000 mg/L <sup>[9]</sup>. Water with a few thousands mg/L of dissolved minerals is classified as lightly saline, but is sometimes used in area where less mineralized water is available <sup>[9]</sup>. Water from some wells and springs can contain very large concentrations of dissolved minerals and waste, which are harmful or dangerous to humans, animals and plants. The quality of water resources in the study area can be degraded by excessive waste disposal and dissolved minerals which can upsets the balance that exist between plants and animals with severe effects on all forms of life.

# Carbonate (HC0<sub>3</sub><sup>-</sup>)

The WHO standard has given the guideline maximum concentration of carbonate to be 50 mg/L<sup>[10]</sup>. The determined concentration of the carbonate in the surface water range between 19.76 mg/L and 66.15 mg/L, averaging 43.388 mg/L. However, the results of the groundwater (hand dug wells and boreholes) concentration range from 59.30 mg/L to 251.72 mg/L, averaging 116.13 mg/L. The analysis of surface water concentration shows that three of the four water samples analyzed are within the WHO standard limit and is therefore safe, while the ground water concentration for HC0<sub>3</sub><sup>-</sup> is higher than the WHO standard limit. This is perhaps hard water, which is not safe or suitable for drinking, except by boiling.

# Nitrate (N03<sup>-</sup>) and sulphates

The WHO standard has shown that the guideline maximum concentration for nitrate is 50 mg/L<sup>[10]</sup>. The result of the analysis shows that the concentration of nitrate in the surface water is vary from 0.02 mg/L up to 15.94 mg/L, with an average concentration of about 5.19 mg/L. On the other hand, the results of the ground water shows concentration ranging between, 0.016 mg/L and 0.39 mg/L, averaging 0.10 mg/L. The surface and ground water concentrations fall below the WHO standard limit and are therefore safe for drinking. The low concentration of nitrate in the water samples might be that the water contains less or no organic matter <sup>[2]</sup>. The WHO standard for the guideline maximum concentration of sulphate ion is 250 mg/L [<sup>10]</sup>. The concentrations of sulphate in surface water varies from 0.25 mg/L up to 5.32 mg/L, averaging 3.30 mg/L. Conversely, the concentration of sulphates in the ground water ranges from 0.25 mg/L to 0.66 mg/L. The surface and ground water concentration fall below the WHO standard limit and sreater and ground water concentration fall below the ground water ranges from 0.25 mg/L to 0.66 mg/L. The surface and ground water concentration fall below the WHO standard limit and is therefore safe for drinking.

# Chloride (Cl<sup>-</sup>)

The WHO standard has shown the guideline maximum concentration for chloride as 250 mg/L <sup>[10]</sup>. The results of the analysis show that the concentrations of chloride ion in the surface water range between 6.62 mg/L, and 29.78 mg/L with an average concentration of about 15.48 mg/L. Alternatively, the concentration of chloride in the ground water varies from 16.54 mg/L to 25.52 mg/L, averaging 21.27 mg/L, which falls below the WHO standard limit and are therefore is safe for drinking.

# Calcium (Ca<sup>2+</sup>)

The WHO standard showed that the guideline maximum concentration for calcium is 100 mg/L <sup>[10]</sup>. The results of the analysis carried out on the samples collected from the surface water resources shows that the concentrations of calcium to be 4.38 mg/L, 4.21 mg/L and 3.64 mg/L in rain water, pond water and stream water, respectively. This concentration is below the maximum concentration guideline of the WHO <sup>[10]</sup> standard for drinking water, meaning that the surface water is safe for drinking. Similarly, the concentration of calcium in ground water ranges from 1.48mg/L to 4.120 mg/L, averaging 2.89 mg/L. These values are below the WHO standard limit and thus are consider to be safe for drinking.

# Magnesium (Mg<sup>2+</sup>)

The WHO standard showed that the guideline maximum concentration for magnesium (Mg) is 50 mg/L <sup>[10]</sup>. The results of the analysis on the surface water samples show magnesium concentrations to 2.64 mg/L, 2.50 mg/L, 2.49 mg/L and 2.10 mg/L in RW, PW1, PW2 and SW, respectively. These concentrations are below the maximum concentration guideline of the WHO <sup>[10]</sup> standards for drinking water, meaning that the surface water are safe for drinking. However, the results for the ground water (hand dug wells and boreholes) concentrations ranges from 1.21 mg/L to 2.53 mg/L, which are the below the WHO standard limit and is said to be safe for drinking.

### Potassium (K<sup>+</sup>)

The WHO standard for the guidelines maximum concentration for potassium is 12.00 mg/L <sup>[10]</sup>. The concentrations of potassium in the varies between 3.04 mg/L and 4.11mg/L for the surface, and between 2.04 mg/L and 3.73 mg/L for the groundwater. These values are below the WHO <sup>[10]</sup> recommended permissible limit of the concentration of potassium in drinking water. Therefore, the water are considered safe for drinking.

### Sodium (Na<sup>+</sup>)

The WHO standard for the guidelines maximum concentration for sodium is 200.00 mg/L <sup>[10]</sup>. The concentrations of sodium in the range from 0.13 mg/L to 0.78 mg/L for the surface, and varies between 0.13 mg/L and 4.76 mg/L for the groundwater. These values are below the WHO <sup>[10]</sup> standard limit and are therefore safe for drinking.

### Lead (Pb<sup>2+</sup>)

The WHO shows that the standard guideline maximum concentration for lead is 0.010 mg/L and the maximum admissible concentration is to be 0.005 mg/L <sup>[10]</sup>. The results of the analysis carried out on samples collected from the surface water shows that the concentrations of lead varies between 0.031mg/L and 0.30 mg/L, averaging 0.12 mg/L. These values concentration are above the WHO standard limit for drinking water, hence it is suitable for drinking. On the other hand, the concentration of lead (Pb) in the groundwater ranges from 0.031mg/L up to 0.046mg/L, which is also higher than the WHO recommended value. This shows that both the surface and ground water may not be safe or suitable for drinking.

# Cadmium (Cd<sup>2+</sup>)

The WHO standard showed that the guideline maximum concentration for cadmium (Cd) is 0.003 mg/L and the maximum admissible concentration to be 0.005 mg/L. The concentrations of cadmium range between 0.004 mg/L and 0.075 mg/L, averaging 0.054 mg/L for the surface water. Likewise, the concentrations of cadmium (Cd) in the ground water varies from 0.011mg/l to 0.06mg/l, averaging 0.036 mg/L. The concentration of cadmium (Cd) in both surface and ground water resources are higher than the WHO permissible limit. This showed that the surface water and the ground water in the study area may not be safe for drinking.

### Zinc (Zn<sup>2+</sup>)

The WHO standard for the guidelines maximum concentration for sodium is 3.00 mg/L <sup>[10]</sup>. The concentrations of sodium in the range from 0.13 mg/L to 0.22 mg/L for the surface, and varies between 0.10 mg/L and 0.12 mg/L for the groundwater. These values are below the WHO<sup>10</sup> standard limit and are therefore safe for drinking.

### Nickel (Ni<sup>2+</sup>)

The WHO standard shows that the guideline maximum concentration for nickel (Ni) is 0.020mg/L <sup>[10]</sup>. The results of the analysis from the surface water resources show that the concentrations Nickel in the water surface water samples range from 0.06 mg/L to 0.35mg/L, averaging 0.133mg/L. The concentration of nickel is above the WHO for the surface water. However, the concentration of nickel in the ground water ranges from 0.030 mg/L to 0.045 mg/L. These concentrations in the surface and groundwater are higher than WHO permissible limit and may not be safe for human consumption.

The high concentration of the ions in the water may be as a result of domestic waste disposal along the water, the use of chemicals for weeds control, the use of chemical for agricultural produce and also from the underlying rocks of the area. Water as a solvent is capable of dissolving the minerals of the underlying rocks and wastes which in turn percolates into the ground as contaminants to the ground water of the area, while others flows into the surface as run-off into streams and ponds in the area. Moreso, with the increase in improper waste disposal, use of chemicals in controlling weeds and fertilizers usage without taking into consideration the environmental effects on both ground and surface water resources of the area, may pollute the water resources of this area thereby making it unfit for human consumption.

### Iron (Fe)

The WHO standard showed that the guideline maximum concentration for iron (Fe) is 0.30 mg/L and the maximum admissible concentration for iron is 0.20 mg/L <sup>[10]</sup>. The results of the analysis carried out on the samples collected from the surface water for iron shows concentration of iron varying between 0.24 mg/L and 0.74 mg/L, with an average concentration of about 0.41 mg/L. The groundwater concentration ranges from 0.12 mg/L to 0.19 mg/L, averaging 0.17 mg/L. The studied surface and ground water resources show that seven of the ten water samples analyzed for iron is within the WHO <sup>[10]</sup> permissible limit. Only the rainwater, stream water and hand pump well water have values of 0.74 mg/L 0.42 mg/L and 0.33 mg/L, respectively. The high level of iron recorded in this study might be due to the natural occurrence of iron in the geological strata of the soil, corrosion of iron and steel materials in the case of rain water hand pump well or leachates from dump sites and vehicles <sup>[2]</sup>.

### Manganese (Mn)

The WHO standard guideline of maximum concentration for manganese (Mn) is 0.05mg/L <sup>[10]</sup>. The results of the analysis from the surface water for manganese are below detective limit except for the rainwater that recorded a value of 0.010 mg/L, averaging 0.003mg/L, while the groundwater is also below dilution limits of 0.00 mg/L. This shows that both concentrations are below the permissible limit by WHO. The water is therefore safe for drinking or human consumption.

### Copper (Cu)

The WHO standard shows that the guideline maximum concentration for copper (Cu) is 0.05 mg/L <sup>[10]</sup>. The concentrations of copper varies from 2.01mg/L to 3.61mg/L, averaging 2.99 mg/L. However, the concentration of copper in the groundwater resources ranges between 0.70mg/L and 2.41mg/L averaging 1.975mg/L. The concentration of copper in both surface and groundwater resources of the area is above the WHO <sup>[10]</sup> permissible limit for

drinking water. The high concentration of copper in the water could be due to sediment dissociation, acid rain or water can also lead to corrosion of copper pipes and copper galvanized roofing sheet and so not safe for drinking.

### pН

The WHO standard limits for pH is 6.5 - 8.5. The results of the analysis from the surface water for pH range between 6.95 and 7.18, averaging 7.04. The groundwater concentration ranges from 6.82 to 7.66, averaging 7.11. The results show that the pH concentration in both surface and groundwater resources are below the WHO <sup>[10]</sup> permissible limits, hence, the water is safe for drinking.

### Total dissolved solid

The WHO standard limit for total dissolved solid (TDS) is 500 mg/L. The results of the analysis of the surface water for TDS shows concentration ranging between 0.00 mg/L to 20.0mg/L, averaging 15.0 mg/L the ground water concentration ranges from 20.0mg/L to 360.0mg/L, averaging 136.67 mg/L. These values are below the WHO <sup>[10]</sup> recommended limit for drinking water, hence both the surface and groundwater are safe for drinking.

### Alkalinity

The WHO standard guideline of maximum concentration for alkalinity is 100 mg/L. The results shows that the surface water has alkalinity varying between 13.33 mg/L and 100.0 mg/L. The concentration of alkalinity in groundwater ranges from 120.0 mg/L to 493.33 mg/L. The surface and ground water resources show that three out of the ten water samples analyzed for alkalinity is within the WHO limits of 100mg/L, With the exception of pond water one, hand dug well water 1-4, hand pump water and motorized borehole water have values of 133.33 mg/L, 206.67 mg/L, 250.0 mg/L, 120.0 mg/L, 130.0 mg/L, 493.33 mg/L and 193.33 mg/L, respectively. The high level of alkalinity recorded in those samples might not be far from the nature of soil that contains high level of HC03<sup>-</sup>, S03<sup>2-</sup> and C03<sup>2-</sup>.

### **Microbiological analysis**

The result of the microbiological analysis shows that the first pond water (PW 1) has 1.10  $\times 10^2$  CFU/100mL for TBC,  $3.0 \times 10^1$  CFU/100mL for Total Coliform (TC) and absent of E. coli (EC). The second pond water (PW 2) has  $1.29 \times 10^2$  CFU/100mL for TBC,  $1.7 \times 10^1$  CFU/100mL for TC and present of EC. The stream water (SW) has  $2.35 \times 10^2$  CFU/100mL for TBC,  $2.3 \times 10^1$  CFU/100mL for TC and present of EC. The sample A (rainwater) has  $5 \times 10$ CFU/100mL for TBC,  $1.0 \times 10^1$ CFU/100mL for TC and absent of EC. The Hand dug well (1) has  $5.0 \times 10^1$ CFU/100mL for TBC which is below WHO limits of 100mL. The TC in the same water sample is  $3.0 \times 10^1$ CFU/100mL for TBC,  $3 \times 10$ CFU/100mL for TC and present of EC. While sample water H (Hand dug well 4) has the TBC below WHO limit, and TC of  $1.0 \times 10^1$ CFU/mL as against 0CFU/100mL of WHO and present of EC. Some of these figures are far and above WHO standard for potable water of 100CFU/100mL for TBC, 0CFU/100mL for TC and EC, respectively. The high microbial load of the water samples analyzed might be due to poor handling, expose of wells, growth of algae, fungi and the nearness of water sources to dumpsites.

# Average distributions of ions

The average value of the cations are 3.50 mg/L, 2.16 mg/L, 3.16 mg/L and 1.07 mg/L for  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$  and  $Na^+$ , respectively. Calcium has the highest rate of distribution as shown in Figure 2. This is followed by potassium, magnesium with sodium been the least. From the distribution of cations in the water resources, it was observed that the concentrations of  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$  and  $Na^+$ , are gradually rising due to the human activities on the water resources. These activities include indiscriminate dumping of refuse, gaseous emission from vehicles and application of fertilizers on farmland. It is also noted that the average concentration of these

cations fall below the value of WHO <sup>[10]</sup> standard for drinking water. Figure 3 shows that the average concentration of hydrogen trioxocarbonate (iv) ion (HC0<sub>3</sub><sup>-</sup>) is very high in the study area, with an average concentration of 80.21 mg/L. This is followed by chloride, nitrate and suphate with average concentrations of about 18.38 mg/L, 2.645 mg/L and 1.85 mg/L, respectively. It is observed that, of all the anions distribution in the water resources, only the bicarbonate ion has an average concentration that is more than the WHO recommended standard of 50.00 mg/L. Figure 4 shows the average concentrations of nickel (Ni), lead (Pb) and cadmium (Cd) are 0.27 mg/L, 0.08 mg/L and 0.05 mg/L, respectively. These values are higher than the WHO recommended values of 0.02 mg/L, 0.01 mg/L and 0.03 mg/L for nickel (Ni), lead (Pb) and cadmium (Cd). Zinc (Zn) which the more abundant metal in the analyzed metals has a concentration of 0.14 mg/L as against the WHO standard of 3.00 mg/L. Copper (Cu) is the most abundant trace element in the water resources and it has a high concentration, averaging 2.483 as against the WHO standard of 0.05 mg/L (Figure 5). Iron is the second most abundant trace element in the water samples with an average concentration of 0.193 mg/L, which is higher than the WHO <sup>[10]</sup> recommended standard of 0.30 mg/L. The least abundant tracement in the water samples is manganese (Mn) with an average concentration of 0.002 mg/L as against the WHO <sup>[10]</sup> standard of 0.05 mg/L. Figure 6 revealed that the average concentration of alkalinity is high in the study area, it has an average concentration of about 175.00 mg/L as against WHO <sup>[10]</sup> of 100.00 mg/L. This is follow by the average concentration of TDS with 88.00 mg/L as against WHO <sup>[10]</sup> of 500.00 mg/L, while that of pH is 7.08 as against the WHO standard of 6.5 - 8.5. However, the average concentration of alkalinity is far higher than the WHO standard. The measured pH and TDS values fall below the WHO <sup>[10]</sup> standards.



Figure 2. Average distribution of cations in the water resources of the study area



Figure 4. Average distribution of heavy metals in the water resources of the study area



Figure 3. Average distribution of anions in the water resources of the study area



Figure 5. Average distribution of trace elements in the water resources of the study area



Figure 6. Average distribution of pH, alkalinity and TDS in the water resources of the study area

### 5. Conclusions

Water is a major necessity of life without which the existence of man on earth will be very difficult, hence the need for the provision of quality potable water. The analysis of the water resources (ground and surface waters) of Agbadu-Bunu Community and its environs was carried out with a view of accessing the water quality by determining the concentrations of cations, anions, trace elements, heavy metals, pH, alkalinity and total dissolved solid distributions as well as the microbial loads contained in the water. Result of the physio-chemical analysis revealed that all the cations (Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup> and Na<sup>+</sup>) conformed to WHO standard limits for drinking water. Also, majority of the anions conformed to WHO standard limits for drinking water except for the concentration of HC03<sup>-</sup> in most of the individual water samples. However, the level of pH and TDS are within the WHO limits for drinking water, while the alkalinity concentration in seven sampled water falls above the WHO limits for potable water. The microbiological analysis carried out to determine the microbial loads in the water samples revealed that most of all the water samples analyzed the area is not fit for drinking. The heavy presence of microbial loads (Total Bacterial Count, Total Coliforms Count and E. Coli) in the sampled water makes it unfit for human consumption except in sample G, I and J where the microbial loads fall below the WHO limits for drinking water. It is therefore, of great concern for both the Government, the Scientist, the water companies and individuals to take into cognizance the drinking water quality of the water used around us.

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To whom correspondence should be addressed: Dr. Christopher Baiyegunhi, Department of Geology and Mining, School of Physical and Mineral Sciences, University of Limpopo, Private Bag X1106, Sovenga 0727, Limpopo Province, South Africa, E-mail: christopher.baiyegunhi@ul.ac.za

# Article

# **Open Access**

Geophysical, Geotechnical and Water Quality Assessment of Gosa Dumpsite in Abuja, Nigeria: Implications for the Future Development of a Sanitary Landfill

Olusegun Omoniyi Ige<sup>1\*</sup>, Christopher Baiyegunhi<sup>2</sup>, Bashiru Ojulari<sup>1</sup>, Olufemi Ogunsanwo<sup>1</sup> and Temitope Love Baiyegunhi<sup>3</sup>

- <sup>1</sup> Department of Geology and Mineral Sciences, University of Ilorin, Private Mail Bag 1515, Ilorin, Kwara State, Nigeria
- <sup>2</sup> Department of Geology and Mining, School of Physical and Mineral Sciences, University of Limpopo, Private Bag X1106, Sovenga 0727, Limpopo Province, South Africa
- <sup>3</sup> Department of Geology, Faculty of Science and Agriculture, University of Fort Hare, Private Bag X1314, Alice, 5700, Eastern Cape Province, South Africa

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#### Abstract

Geophysical, geotechnical and water quality investigations of Gosa dumpsite in Abuja, Nigeria were performed to unravel the feasibility of upgrading it to a modern sanitary landfill. Nineteen vertical electrical soundings were carried out to geoelectrically characterized the site. In addition, eight soil and nine water samples were analyzed to determine their geotechnical and physiochemical properties as well as their usability. The inversion of VES-DC data indicate 3-4 geoelectric layers. Geotechnically, the soils are classified as well graded clayey-sand to silty-sand with permeability between  $5.1 \times 10^{-7}$  m/s and  $1.1 \times 10^{-6}$  m/s, which makes the groundwater prone to leachate pollution. The physicochemical parameter of the water are generally below the WHO recommended value. It could be inferred that only the northern part of the area possess the required geophysical and geotechnical characteristics for upgrade to a sanitary landfill. The high concentration of radioactive elements in the water revealed that the present dumping activities has negatively impacted the groundwater quality.

Keywords: Geophysical; Geotechnical and groundwater; Sanitary landfill; Nigeria.

#### 1. Introduction

Human population is increasing on daily basis, so is the corresponding quantity of waste contending for space with man and its effect impairing the quality of environment <sup>[1]</sup>. The use of waste disposal by landfill is a very common practice, and the ever increasing demand for waste management makes them a very vital part of human existence. Preferred options of solid waste management are waste reduction at source and re-use <sup>[2]</sup>. However, disposal of solid waste in sanitary landfills is now increasingly gaining acceptance in many nations of the world <sup>[3]</sup>. This can be attributed to the fact that landfills serves as a final repository even for waste managed with other waste disposal techniques. Several researchers including Thompson and Zandi <sup>[4]</sup>, Rushbrook <sup>[5]</sup>, and Carra and Cossu <sup>[6]</sup> have documented that solid waste disposal in landfills are still the most economic form of disposal in the vast majority of cases. Thus, landfills will continue to be the most attractive disposal method for solid waste. Depending on location, about 95% of the solid wastes generated worldwide are disposed in landfills [7-8]. Since the implementation of early landfills, steps have been taken to improve their designs and management in order to reduce the impact of solid waste on environment and groundwater resources. Though, the same measures have been used worldwide, underdeveloped and developing countries are still struggling with the final disposal of their solid waste.

Landfills in the form of open and uncontrolled dumpsites are the most common waste disposal systems in many cities and towns in Nigeria. Open dumps are the oldest and most common way of disposing off solid wastes, although in recent years, thousands have been closed and many are still being used. Most of these waste landfills are unsuitably designed and managed as a result of capital investment, hence allowing for environmental pollution in those areas where they are located. Recently, the impact of leachate on groundwater and other water resources have attracted a lot of attention because of its overwhelming environmental significance. Landfill leachates are complex; heavy metal components are undoubtedly the most harmful because of their persistence and toxicity <sup>[9]</sup>. Most heavy metals in sanitary landfills, which have anti-seepage protection measures, are retained in the waste in a complex form <sup>[10]</sup>. The leachate from irregular landfills links directly to groundwater, and pollutes the groundwater as well as the hosting or local soil. Presence of heavy and trace metals in drinking water as a consequence of groundwater contamination by leachates is also a major health and environmental problem in Nigeria. Certain toxic heavy metals like lead, mercury, cadmium, minerals and man-made synthetic chemicals present in wastes may contribute to environmental degradation that leads to poor health, disease or death <sup>[11]</sup>. Waste has been recognized as one of the major problems confronting governments and city planners in Nigeria, thereby posing a serious threat to environmental quality and human health <sup>[12]</sup>.

The quantity and generation rate of solid wastes in Nigeria have increased at an alarming rate over the years with lack of efficient and modern technology for the management of the wastes <sup>[13]</sup>. Generally, the higher the economic development and rate of urbanization, the greater the amount of solid waste produced <sup>[14]</sup>. The rapid population increase due to urbanization in Abuja metropolitan areas (i.e. Gosa village) have caused difficulties for the state and local environmental protection agencies in providing an effective and efficient municipal solid waste management <sup>[15]</sup>. Presently, there is no sanitary landfill in the rapidly growing city of Abuja (Federal Capital of Nigeria). The waste collected from the city are disposed in designated dumpsite which include Mpape (closed), Ajata (operational), Kubwa (closed) and Gosa (operational). These dumpsites are characterized by indiscriminate dumping on ground surface and persistent burning of waste. Waste picking is common at the Gosa dumpsite, which is usually an urban phenomenon <sup>[16]</sup>. Gosa dumpsite is located in Gosa village, Abuja, Nigeria. The management of municipal solid waste has become a major environmental problem, especially for fast growing cities like the current federal capital; FCT Abuja, with generation amount increasing on yearly basis <sup>[17]</sup>. Leachate movement from waste sites or landfills pose a high risk to groundwater resource if not adequately managed. Control of heavy metals in leachates has therefore become the focus of landfill management. A suitable solid waste sanitary landfill site should be characterized by proper hydrological, geological and environmental conditions. For these reasons, waste sanitary landfills must be specially designed, constructed and managed to keep them safe during operation. In order to achieve safer disposal of municipal solid waste in Abuja and its environs, the Abuja Environmental Protection Board <sup>[18]</sup> propose to upgrade the Gosa dumpsite to a modern sanitary landfill. Modern landfills are highly engineered containment systems, designed to minimize the impact of solid waste (refuse, trash, and garbage) on the environment and human health [19]. In modern landfills, the waste is contained by a liner system <sup>[18]</sup>. The primary purpose of the liner system is to isolate the landfill contents from the environment and therefore, to protect the soil and ground water from pollution originating in the landfill. However, no single geophysical tool can effectively determine the characteristics of a landfill. Integration of geophysical, geotechnical and physiochemical analytical methods provides an important tool in the evaluation and characterization of contaminants or landfills. Pursuant to the above, this study was undertaken to investigate the technical feasibility of upgrading the Gosa dumpsite to a modern sanitary landfill using integrated geotechnical, geophysical and groundwater quality assessment.

### 2. Geology of the area

Abuja is underlain by crystalline basement rocks and the rocks include different textures of granites, coarse to fine, consisting essentially of biotite, feldspars and quartz <sup>[20]</sup>. The rocks in the study area comprises of migmatite gneiss, granodiorite, porhyroblastic gneiss, Pan African granite, granite gneiss and amphibolites (Figure 1). Generally, the North-North East (NNE) and South-SouthWest (SSW) of the FCT are made of gneiss, migmatites and granites



Figure 1. Geological map of the study area <sup>[3]</sup>

which characterize the Northern Nigeria <sup>[21]</sup>. All these rocks have been affected and deformed by the Pan-African thermotectonic event <sup>[22]</sup>. The rocks are highly fractured and jointed showing essentially two fracture patterns, NE-SW and NW-SE<sup>[23]</sup>. These fractures induce structural control on the drainage of Abuja. The out crop of schist belt is found along the Eastern margin of the area and this belt broadens as one moves south wards and maximum size is found to the South Eastern region of the FCT <sup>[21]</sup>. Minor Cretaceous deposits of Nupe sandstones occur in the southern part of FCT between Kwali and Abaji, extending to Rubochi <sup>[23]</sup>. The rocks in the study are have reddish micaceous sandy clay to clay materials, often capped by laterite.

### 3. Materials and methods

#### 3.1. Geophysical investigation

An allied Omega terameter was used to carry out vertical electrical sounding (VES) geophysical survey along five traverses that cut across the study area (Figure 3). Schlumberger array was employed along the NE-SW traverses. A total of nineteen VES stations were used to investigate the area. Sixteen VES stations fall within the study area, while three VES stations (VES 5, VES 16 and VES 19) were taken outside the boundary of the area to extend investigation beyond the dumpsite. The VES data were processed using the software IP2Win developed by Alexey Bobachev, Moscow State University, Russia.

### 3.2. Geotechnical investigation

Eight samples comprising of two undisturbed and six disturbed samples were analysed geotechnically. The two undisturbed samples were recovered from two boreholes drilled during Standard Penetrometer Test (SPT), while the six disturbed samples were recovered from six trial/test pit dug manually. The test pits were dug to depths of 1.0 m and sampling done at 1.0 m. About 40 kg of disturbed soil samples were taken at each test pit. The soil samples recovered from the two boreholes (BH1 and BH2) and the six test or trial pits (T1-T6) were analysed to obtain required geotechnical parameter in landfill characterization. Preliminary geotechnical classification and identification tests such as water content, bulk density, specific gravity, grain size distribution, Atterberg consistency limits, specific gravity, permeability and compaction test were carried out on the samples based on the British Standard (BSI 1377:1990). Each geotechnical test was performed twice on the same soil sample under the same condition in order to determine the reliability of the geotechnical test results.

#### 3.3. Groundwater quality assessment

A total of nine water samples were collected from hand-dug wells, boreholes, springs and river in the study area. A clean 1.0 litre plastic container was filled with water sample from each sampling location after rinsing it with the same water sample in each case. The filled plastics were immediately capped tight to prevent the trapping and/or absorption of atmospheric oxygen. The samples were adequately labelled, refrigerated and sent to the laboratory to prevent cationic adsorption on the wall of the container during storage. Physicochemical analysis of the water samples were performed at Center for Energy Research and Development

(CERD), Obafemi Awolowo University, Ile-Ife, Osun State, Nigeria. Cationic and anionic analyses were carried out using Atomic Adsorption Spectrometry (AAS) in order to determine the concentration of  $Cd^{2+}$ ,  $Cr^{2+}$ ,  $Fe^{2+}$ ,  $Cu^{2+}$ ,  $Pb^{2+}$ ,  $Ni^{2+}$ ,  $Zn^{2+}$ ,  $As^{3+}$ ,  $K^+$ ,  $Na^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Cl^-$ ,  $NO_3^-$ ,  $HCO_3^-$ ,  $CO_3^{2-}$ ,  $PO_4^{2-}$  and  $SO_4^{2-}$ . In addition, physical parameters like pH, colour, temperature, total dissolved solid (TDS), turbidity, electrical conductivities, total hardness and total alkalinity were also determined.

#### 4. Results and discussion

#### 4.1. Geophysical assessment

The results of geophysical investigation is summarized in Table 1. The 2D pseudosection for the VES data along traverses 1-5 is shown in Figure 2.



Figure 2. Map showing the study area as well as the VES, borehole, trial pits and groundwater sampling points Table 1. Summary of the geophysical characteristics of the study area

Traverse	VES Sta- tion	No of Layers	Apparent Resistivity (Ωm)	Thickness (m)	Depth to Basement (m)
	1	4	796/1883/903/4455	1.45/8.38/17.4/	27.2
	2	4	120/522/685/1000	1.18/6.57/16.6/	24.4
1	3	4	74.2/415/755/2663	1.19/4.92/15.9/	22.0
	4	4	742/1474/590/3026	1.28/5.17/13.4/	19.8
	5	4	916/3871/1579/4780	1.47/11.8/8.38/	21.6
	6	4	613/3190/1018/4249	1.0/6.99/15.7/	23.7
2	7	4	461/850/554/5652	1.88/7.72/11.7/	21.3
2	8	4	231/1503/554/4080	1.13/8.09/12.0/	21.2
	9	4	490/1699/922/6790	1.13/2.79/15.3/	19.2
	10	3	5426/2040/8849	2.31/15.2/	17.5
3	11	3	356/478/1000	0.5/6.05/	6.6
	12	3	686/416/1570	2.17/4.76/	6.9
	13	4	2945/408/1818/4455	1.08/4.92/8.72	14.7
4	14	3	1519/3334/7682	2.1/3.49/	5.6
	15	3	2659/1279/7483	1.02/3.51/	4.5
5	16	3	752/2353/5599	2.17/3.94	6.1
	17	3	3799/1264/6453	0.79/6.58	7.4
	18	3	1000/2259/5427	1.18/4.08	5.3
	19 3		110/736/5934	0.13/7.0/	7.1

Four geologic layers were delineated at VES 1-VES 9 and VES 13, whereas, three geologic layer were delineated at VES 10, and VES 12-VES 19. These four layers correspond to lateritic top soil, laterites/clayey sand, weathered basement and fresh basement, respectively. The weathered basement thinned out at VES 10 and VES 12-VES 19 where outcrop of the basement occurred as low lying hill. The resistivity value of the lateritic top soil varies from 110  $\Omega$ m at VES 19 to 5426  $\Omega$ m at VES 10, while its thickness varies from 0.5 m - 2.31 m at VES 10 and VES 11. The second layer has resistivity value ranging from 408  $\Omega$ m at VES 13 to 3871  $\Omega$ m at VES 5. In addition, the thickness of the second layer varies from 2.79 m at VES 9 to 15.2 m at VES 10. This layer is clayey and therefore more significant in providing material barrier/seal in the proposed sanitary landfill. The 2D resistivity pseudo sections (Figure 3) show that this layer thinned out towards the south of the study area. Furthermore, depth to basement also becomes shallower southerly from a maximum of 27.2 at VES 1 to a minimum of 4.5 m at VES 15. Therefore, the required clay/clayey material required as landfill liner is more abundant in the Northern part of the study area. The third layer is the weathered basement, which directly overlies the fresh basement. Its resistivity value varies from 416  $\Omega$ m at VES 12 to 3334  $\Omega$ m at VES 14, whereas its thickness varies from 3.49 m at VES 14 to 17.4 m at VES 1. The last layer corresponds to the basement and its resistivity value varies from 1000  $\Omega m$  at VES 2 and VES 11 to 8849  $\Omega m$  at VES 10. The relatively high resistivity values are possibly due to reworking of the soil surface or presence of compacted lateritic hard pan.



Figure 3. 2D resistivity pseudosection along traverse 1 (VES 1-5), traverse 2 (VES 6-9), traverse 3 (VES 10-12), traverse 4 (VES 13-15) and traverse 5 (VES 16-19)

# 4.2. Geotechnical assessment

The results of geotechnical analysis is summarized in Table 2. The geotechnical requirements for soil to be used as landfill liners as recommended and confirmed by Ige et al.<sup>[24]</sup> is presented in Table 3.
Comple	WC	BD	SG		Attachang Limit	Compactio	on Test	Permeability,
Sample	(%)	(kg/m <sup>3</sup> )		USC	Atterberg Limit	Standard	Modified	k (m/sec)
Т1	1.3	1.28	2.5 5	SC- SM	LL= 39%; PL= 21%; Ip = 21%; FI= ; TI= AC=0.33 ; SL=	OMC = 16 %; MDD =1.8g/cm <sup>3</sup>	OMC = 14 %; MDD =1.83g/cm <sup>3</sup>	4.892 x 10 <sup>-7</sup>
T2	2.8	1.26	2.6 4	SC- SM	LL= 35%; PL= 21%; Ip =14% ; FI=22 ; TI=0.64; AC=0.28 ; SL=	OMC =16.5 %; MDD =1.73g/cm <sup>3</sup>	OMC = 16 %; MDD =1.78g/cm <sup>3</sup>	4.467 x 10 <sup>-7</sup>
Т3	0.6	1.66	2.6 0	SP		OMC = 10 %; MDD =1.8g/cm <sup>3</sup>	OMC = 9 %; MDD =1.83g/cm <sup>3</sup>	1.361 x 10 <sup>-6</sup>
T4	3.7	1.38	2.6 6	SC- SM	LL=30% ; PL=18% ; Ip =21% ; FI= ; TI= AC= 0.33; SL=	OMC = 15.5 %; MDD =1.8g/cm <sup>3</sup>	OMC = 14.5%; MDD =1.85g/cm <sup>3</sup>	6.080 x 10 <sup>-7</sup>
Т5	2.1	1.65	2.6 5	SC- SM	LL=30% ; PL=17%; Ip =13% ; FI= 25; TI= 0.5; AC=0.6 ; SL=	OMC =12 %; MDD =1.79g/cm <sup>3</sup>	OMC = 11 %; MDD =1.82g/cm <sup>3</sup>	1.101 x 10 <sup>-6</sup>
Т6	5.9	1.35	2.6 0	SC- SM	LL= 34%; PL=23% ; Ip =11% ; FI= 25; TI= 0.6; AC=0.2 ; SL=	OMC = 16 %; MDD =1.7g/cm <sup>3</sup>	OMC = 14 %; MDD =1.76g/cm <sup>3</sup>	5.142 x 10 <sup>-7</sup>
BH1	14. 6	1.82	2.5 5	SC	LL= 24%; PL=16% ; Ip = 7%; FI= ; TI= AC= ; SL=	OMC = ; MDD =	OMC = ; MDD =	
BH2	11	1.85	2.6 4	SC	LL=28% ; PL=17% ; Ip = 11%; FI= ; TI= AC= ; SL=	OMC = ; MDD =	OMC = ; MDD =	

Table 2. Summary of results obtained from geotechnical analysis of the samples

*WC* = Moisture content; *BD* = Bulk density; *S.G* = Specific gravity; *USC* = Unified Soil Classification; *SP* = Poorly graded sand; *SC* = Well graded clayed-sand; *SM* = Silty-sand; *LL* = Liquid limit; *PL* = Plastic limit; *Ip* = Plastic index; *FI* = Flow index; *TI* = Toughness index; *AC* = Activity of clay; *OMC* = Optimum moisture content; *MDD* = Maximum dry density.

The result of specific gravity mainly indicate values ranging from 2.50 up to 2.69. Based on the classification scheme of Bowles <sup>[25]</sup>, it could be inferred that the soil samples are composed mainly of sand, silt and clay. In addition, based on specific gravity of common minerals given by Das <sup>[26]</sup>, it can be inferred that the dominant minerals in samples are kaolinite and quartz. Kaolinite is non-expansive clay and is generally good for liner materials in dams and landfills. Grain size analysis of soil samples T1, T2, T3, T4 and T6 shows abundance of gravel, sand, silt and are as follows: T1 (0 % Gravel, 50 % sand, 30 % silt, 20 % clay); T2 (25 % Gravel, 45 % sand, 23 % silt, 7 % clay); T3 (10 % Gravel, 90 % sand, 0 % silt, 0 % clay); T4 (10 % Gravel, 60 % sand, 19 % silt, 11 % clay); T5 (25 % Gravel, 57 % sand, 13 % silt, 5 % clay); and T6 (1.2 % Gravel, 48.8 % sand, 36 % silt, 14 % clay).

The grain size analysis shows that the samples are composed predominantly of sand size particles with some amount of silt and clay. The only exception is sample T3 which is composed almost entirely (90 %) of sand size particles had no silt. All the soil samples (except sample T3) are classified as clayed-sand to silty sand (SC-SM) base on Unified soil classification. Sample T3 is classified as poorly graded sand (SP) which implies that all the grain sizes are not well represented. The percentage of sand predominates, follow by silt and sand respectively. The bored samples (BH1 and BH2) can be classified as clayed-sand lateritic soil with dominant percentage of clay (14-30 %). The largest grain size in soil samples is 16 mm and thus satisfies the grain size requirement of  $\leq$  63mm suggested by ONORMS<sup>34</sup> and the values of between 30 mm and 50 mm suggested by Daniel (1993). The percentage of fines contained in the soils are T1 = 50 %, T2 = 30 %, T3 = 0 %, T4 = 30 %, T5 = 18 %, T6 = 50 %, BH1 = 29-30 %, BH2 = 30 %. The values of the percentage of fines also satisfy the required  $\geq$  15 %. All the studied samples have gravel percentages less than the recommendation. This shows

that the soil samples (except T3) are cohesive soil with very little porosity, which is desirable for liner materials.

Parameters	Author(s)	Recommendations	This study
Grain size	Oelzschner <sup>[30]</sup> Bagchi <sup>[31]</sup> ONORMS <sup>[32]</sup> Daniel <sup>[33]</sup> ; Rowe <i>et al.</i> , <sup>[34]</sup>	Clay fraction < 20 % Largest grain size $\leq$ 63mm Silt/clay fraction $\geq$ 15 % Largest grain size < 25mm, %Gravel < 30, % fine $\geq$ 30 %	Clay = $5 - 30 \%$ Largest grain size = 16mm 20 - 50 % fines $\leq 10 \%$ gravel
Atterberg consistency limits	Daniel <sup>[33]</sup> ; Rowe <i>et al.</i> , <sup>[34]</sup> Seymour and Peacock <sup>[35]</sup> Oelzschner <sup>[30]</sup>	$\begin{array}{l} LL \geq 30\%, \ IP \geq 15 \ \% \\ LL \geq 30\%, \ IP \geq 10 \ \% \\ LL \geq 30\%, \ IP \geq 15 \ \% \\ LL \geq 25\%, \ IP \geq 15 \ \% \\ LL \geq 30\%, \ IP \geq 15 \ \% \\ IL \geq 30\%, \ IP \geq 15 \ \% \\ Inorganic \ clay \ of \ low-medium \\ plasticity \ (CL-CI) \ and \\ Ac \ of \ < 1.25 \end{array}$	LL = 22 - 39 %, IP = 5.4 - 18.4 % Ac = 0.2 - 0.6
Moisture content- density relationship	ONORMS <sup>[32]</sup> Taha and Kabir <sup>[29]</sup>	$ \begin{array}{l} \text{MDD} \geq 1.71 \text{ t/m}^3 \\ \text{MDD} \geq 1.71 \text{ t/m}^3 \end{array} $	SP = 1.7 - 1.80 g/cm <sup>3</sup> MP = 1.7683 g/cm <sup>3</sup>
Permeability	Murphy and Garwell <sup>[36]</sup> Mark <sup>[37]</sup> Joyce <sup>[38]</sup> Fred and Anne <sup>[39]</sup>	$ \leq 1 \times 10^{-9} \text{ m/s} \\ \leq 1 \times 10^{-9} \text{ m/s} \\ \leq 1 \times 10^{-8} \text{ m/s} \\ \leq 1 \times 10^{-9} \text{ m/s} $	$5.1 \times 10^{-7}$ m/s - 1.1 × 10 <sup>-6</sup> m/s

Table 3	Comparison	of	geotechnical	criteria ł	hν	different	researchers	with	this	study
Table 5.	Companson	UI	yeotecimicar	Cincenta L	Jy	unierent	researchers	WILLI	uns	Study

The results of the Atterberg consistency limits for samples T1 - T6 (excluding T3) and the bored samples BH1 - BH2 shows that the liquid limits, flow index and toughness index range from 25-39%, 3.7-29.1 and 0.3-0.6, respectively. Plot on the plasticity chart shows that samples T1, T2, T4, T5 and T6 are dominantly of low plasticity (PI: 10-20%), whereas the bored samples (BH1-BH2) are of low to intermediate plasticity (PI < 10 %). Plot on the plasticity chart also confirmed that the excavated samples are composed of inorganic clay of medium plasticity, while the bored samples are of inorganic clay of low plasticity. The clay activity (AC) of soil sample T1, T2, T4, T5 and T6 are 0.33, 0.28, 0.35, 0.6 and 0.2, respectively. Therefore, the samples are composed of inactive clays (Ac < 0.75) (non-swelling clay minerals) such as kaolinite, muscovite. Plot of the soil samples on the Casagrande plasticity chart shows that sample BH1 and BH 2 are inorganic clay of low plasticity (CL), whereas T1-T6 are inorganic clay of intermediate plasticity (CI). The results obtained for clay activity of the soil samples reveals that clay activity in the range of 0.20-0.6, which also support or confirm the dominance of kaolinite clay type. Therefore, the soil samples are recommended for use as landfill liners.

Compaction test was carried out to determine the response of the soil samples to compaction effort. It evaluates the ease with which the soil samples from the area can be improved by compaction. Improvement by compaction is necessary to obtain low hydraulic conductivity, which is desirable in landfill barrier. The soil samples were compacted using both standard proctor and modified proctor methods. Summary of results compaction test of the soil samples are presented in Table 2. The results of the permeability test of sample T1-T6 shows that the permeability values range between  $5.1 \times 10^{-7}$  m/s and  $1.1 \times 10^{-6}$  m/s. The samples needed to be improved by the addition of little amount of clay to meet up to  $1 \times 10^{-8}$  m/s recommended by Ige *et al.* <sup>[1]</sup>. The maximum dry density (MDD) range from 1.7 g/cm<sup>3</sup> up to 1.80 g/cm<sup>3</sup> for samples compacted at both standard and modified proctor. This range satisfy the minimum requirement of 1.7 g/m<sup>3</sup> proposed by Ige *et al.* <sup>[1]</sup> for samples under standard proctor. The MDD values under modified proctor range from 1.76 g/cm<sup>3</sup> to 1.83 g/cm<sup>3</sup> much better than 1.45 g/cm<sup>3</sup> (standard proctor) and 1.64 g/cm<sup>3</sup> (modified proctor) specified by Taha and Kabir <sup>[29]</sup> for soils produced from basement complex rocks to be usable as liners in landfill.

#### 4.3. Groundwater quality assessment

The results of the physicochemical analysis of groundwater samples and the drinking water quality standard, as recommended by World Health Organization (WHO) is summarized in Table 4. The result shows that the pH values for the samples range from 8.1 - 9.4, which is higher than the 6.5 - 8.5 pH values recommended by WHO for drinking water. The colour of drinking water does not have direct implication on human health, nonetheless it may affect the acceptability and aesthetic value of the water <sup>[27]</sup>. Based on WHO recommendation, the colour of drinking water should be less than 15 TCU (True colour unit). The colour range for water samples in the study area range from 1 – 90 TCU. Water samples 1, 2, 3, 4, 6, and 8 have colour range of less than 15 TCU, but water samples 5, 7 and 9 have colour units of 90, 25 and 23, respectively and are therefore not suitable for drinking. The palatability of water with total dissolved solids (TDS) level of less than about 600mg/L is generally consider to be good. Drinking water becomes significantly and increasing unpalatable as TDS level become greater than 1000 mg/L <sup>[28]</sup>. TDS in drinking water originate from natural sources, sewage, urban runoff and industrial waste water. All the water samples have TDS value below 600 -1000 mg/L, which is the WHO recommended value. Turbidity is an important indicator of possible presence of contaminants. Turbidity in water is caused by suspended particles or colloidal matters that obstruct light transmission through the water <sup>[27]</sup>. The WHO recommended turbidity of small water supply to be at least less than 5 NTU (Nephelometric turbidity unit). The turbidity of water samples 1, 2, 3, 5, 6 and 8 are below the WHO <sup>[27-28]</sup> recommended value. However, water samples 5, 7, and 9 have turbidity of 74.64 NTU, 8.31 NTU and 7.43 NTU, respectively, which are above the WHO <sup>[27]</sup> recommended value.

According to the Standard Organization of Nigeria (SON) <sup>[29]</sup>, the acceptability threshold value for drinking water conductivity is  $\leq 1000 \,\mu$ S/cm. The conductivity of water samples range from 722.56–1855.02 µS/cm. Water samples 3, 8, and 9 have electrical conductivities of approximately 1199  $\mu$ S/cm, 1855  $\mu$ S/cm and 1766  $\mu$ S/cm, respectively. These values are higher than the SON <sup>[28]</sup> recommended value, hence the water is unfit for drinking. Furthermore, the most significant water quality guideline on crop productivity is the water salinity hazard as measured by electrical conductivity <sup>[24]</sup>. Very high electrical conductivity can lead physiological drought in plants. Todd <sup>[26]</sup> classified water having electrical conductivity between 750  $-2000 \ \mu$ S/cm as permissible for irrigation. Thus, the water in the study area is permissible for irrigation purpose. Hardness is the traditional measure of the capacity of water to react with soap, hard water requiring considerably more soap to produce lather <sup>[27]</sup>. It is usually predominantly caused by the presence of dissolved calcium (Ca<sup>2+</sup>) and magnesium (Mq<sup>2+</sup>) cations. Hardness in drinking water affects its acceptability (taste) and a taste threshold of 100–300 mg/L is established for calcium. In addition, water with hardness above 200 mg/L may cause scale deposition in distribution and storage systems. High soap consumption and scum formation are major negative impact of hardness in water. Total hardness of water samples vary from 40-232 mg/L of CaCO<sub>3</sub>. Therefore, all the water samples from the study area have total hardness within the WHO recommended value. Total Alkalinity is important factor in the prevent ion of water quality problem associated with corrosion of iron pipes (either cast or ductile) use in distribution system. The corrosion of ion in distribution system usually result in degraded water quality commonly called "red water". Successful control of ion corrosion has been achieved by adjusting the pH to the range 6.8 – 7.3, hardness and alkalinity to at least 40 mg/L (as calcium carbonate). However, the annual mean of alkalinity of water sample from the area is 45.50 mg/L CaCO<sub>3</sub> and thus satisfy the WHO <sup>[27]</sup> recommended value of total alkalinity of at least 40 mg/L (as CaCO<sub>3</sub>) for the prevention of "red water".

					WHO (20	)11) Standard
				Std	Health-	Acceptability
Measured Parameter	Min.	Max.	Mean	Deviation	based	Threshold/
				Deviation	Guideline	Optimum
					Value	Value
pH	8.10	9.40	8.72	0.40		6.5-8.5
Colour (Pt/Co Units) Raw	1.00	90.00	17.67	28.66		<15 TCU
Temperature (°C)	37.80	39.30	38.28	0.44		
Total Dissolved Solid (mg/l)	244.84	398.33	319.72	55.85		1000
Turbidity (NTU)	1.17	74.64	11.40	23.86		< 5 NTU
Electrical Conductivities (µS/cm)	722.56	1855.02	1106.39	422.18		1000
Total Hardness (mg/l CaCO <sub>3</sub> )	40.00	232.00	127.11	60.82		100-300
Total Alkalinity (mg/l CaCO <sub>3</sub> )	32.00	302.00	130.67	87.61		At least 40
$Cd^{2+}$ (mg/l)	0.001	0.017	0.009	0.005	0.003	
$Cr^{2+}$ (mg/l)	0.004	0.023	0.015	0.007	0.05	
$Fe^{2+}(mg/l)$	0.003	0.029	0.018	0.009		
$Cu^{2+}(mg/l)$	0.003	0.128	0.059	0.045	2	
$Pb^{2+}(mg/l)$	0.000	0.018	0.008	0.006	0.01	
Ni <sup>2+</sup> (mg/l)	0.002	0.140	0.073	0.050	0.07	
$Zn^{2+}(mg/l)$	0.004	0.161	0.084	0.054	3	
$As^{3+}$ (mg/l)	0.000	0.020	0.009	0.007	0.01	
K <sup>+</sup> (mg/l)	0.054	0.322	0.184	0.091		
Na <sup>+</sup> (mg/l)	0.084	0.721	0.364	0.239		200
Ca <sup>2+</sup> (mg/l)	0.052	0.131	0.096	0.027		
$Mg^{2+}(mg/l)$	0.013	0.089	0.057	0.032		
Cl <sup>-</sup> (mg/l)	15.50	23.01	20.39	2.81		200 - 300
NO <sub>3</sub> <sup>-</sup> (mg/l)	3.28	12.27	8.77	3.31		
HCO <sub>3</sub> <sup>-</sup> (mg/l)	36.00	238.00	115.22	60.31		
$CO_3^{2-}$ (mg/l)	8.00	64.00	44.00	26.53		
$PO_4^{2-}(mg/l)$	0.21	1.52	0.74	0.49		
$SO_4^{2-}$ (mg/l)	14.43	31.19	20.95	5.85		

Table 4. Summary of physiochemical parameter in the studied samples and WHO Standard	d [27-	27	7	.7	9	2	2	2	2	2	C	[	C	C	C	ſ	C	C	[	C	¢,	C	C	C	C	C	¢,	Ç	2	(	7	2	7	2	7	2	2	2	2	2	(	C	C	C	l	
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In epidemiological studies, an association has been found between exposure to chromium (VI) by the inhalation route and lung cancer <sup>[27]</sup>. The WHO recommended maximum value for chromium in drinking water is 0.05 mg/L. The chromium concentration in water range from 0.004–0.023 mg/L. Therefore, all the water samples from the study area are free from chromium contamination. Copper is both an essential nutrient and a drinking-water contaminant <sup>[28]</sup>. Studies are ongoing on the long-term effects of copper on sensitive populations, such as carriers of the gene for Wilson disease and other metabolic disorders of copper homeostasis. The WHO <sup>[27]</sup> recommended maximum value for copper in drinking water is 2 mg/L. The copper concentration in water samples range from 0.003–0.128 mg/L. Therefore, all the water samples are free from copper contamination. Exposure to lead is associated with a wide range of effects, including various neurodevelopmental effects, mortality (mainly due to cardiovascular diseases), impaired renal function, hypertension, impaired fertility and adverse pregnancy outcomes <sup>[28]</sup>. The amount of lead dissolved from the plumbing system depends on several factors, including pH, temperature, hardness and standing time of water, with soft, acidic water being the most plumbosolvent. The WHO recommended maximum value for lead in drinking water is 0.010 mg/L. The lead concentration in water samples range from 0.001-0.018 mg/L. Samples 6, 8 and 9 have arsenic concentration of 0.011 mg/L, 0.015 mg/L and 0.018 mg/L, respectively and are therefore lead-contaminated. Smoking and occupational exposure are the two main sources of Nickel in the population while water is generally a minor contributor to the total daily oral intake. Allergic contact dermatitis is the most prevalent effect of nickel in the general population <sup>[27]</sup>. The International Agency for Research on Cancer (IARC) documented that inhaled nickel compounds are carcinogenic to humans (Group 1) and that metallic nickel is possibly carcinogenic (Group 2B). The WHO recommended maximum value for nickel in drinking water is 0.070 mg/L. The Nickel concentration in the water samples range from 0.002–0.140 mg/L. Samples 5, 6, 7, 8 and 9 have nickel concentration of 0.101 mg/L, 0.096 mg/L, 0.094 mg/L, 0.0127 mg/L and 0.140 mg/L, respectively and are therefore nickel-contaminated. Zinc is an essential trace element found in virtually all food and potable water in the form of salts or organic complexes. However, drinking-water containing zinc at levels above 3 mg/L may not be acceptable to consumers <sup>[27]</sup>. The WHO recommended maximum value for zinc in drinking water is 3 mg/L. The zinc concentration in water samples range from 0.004 – 0.161 mg/L. Hence, all the water samples from the area are free from zinc contamination.

The International Programme on Chemical Safety (IPCS) reported that long-term exposure to arsenic in drinking-water is causally related to increased risks of cancer in the skin, lungs, bladder and kidney <sup>[27]</sup>. The WHO recommended maximum value for arsenic in drinking water is 0.010 mg/L. Arsenic concentration in the water samples range from 0.001 – 0.020 mg/L. Samples 5, 8 and 9 have arsenic concentration of 0.012 mg/L, 0.017 mg/L and 0.020 mg/L, respectively and are therefore arsenic-contaminated. Source of calcium in underground water is usually weathering of parent rock materials especially in limestone region. Calcium ion is a major cause of hardness in drinking water and thus affects the acceptability and aesthetic value of water. The taste threshold for the calcium ion is in the range of 100 - 300 mg/L, depending on the associated anion <sup>[27]</sup>. The collected water samples have calcium concentration ranging from 0.052 – 0.131 mg/L and thus satisfy the WHO taste threshold value. Magnesium ions contribute to hardness in water and the presence of magnesium ion in drinking water is prevalent in gypsum region. Drinking-water in which both magnesium and sulphate are present at high concentrations can have a laxative effect, although data suggest that consumers adapt to these levels as exposures continue <sup>[28]</sup>. The water samples have magnesium ion concentration ranging from 0.013–0.089 mg/L and thus satisfy the WHO recommendation <sup>[27]</sup>.

The main source of human exposure to chlorine is the addition of salt to food, and the intake from this source is usually greatly in excess of that from drinking-water <sup>[27]</sup>. Other source of chloride in drinking-water include sewage and industrial effluents, urban runoff containing de-icing salt and saline intrusion. No health-based guideline value is proposed for chloride in drinking-water. However, chloride concentrations in excess of about 250 mg/L can give rise to detectable taste in water and excessive chloride concentrations increase rates of corrosion of metals in the distribution system, depending on the alkalinity of the water <sup>[27]</sup>. This can lead to increased concentrations of metals in the supply. The range of concentration of chloride in water sample is between 15.50-23.01 mg/L. These values falls below the WHO recommended maximum value range of 200-300mg/L and thus safe and acceptable for drinking. Nitrate ( $NO_3^{-}$ ) is found naturally in the environment and is an important plant nutrient. It is present at varying concentrations in all plants and is a part of the nitrogen cycle. Nitrate can reach both surface water and groundwater as a consequence of agricultural activity (including excess application of inorganic nitrogenous fertilizers and manures), from wastewater disposal and from oxidation of nitrogenous waste products in human and animal excreta, including septic tanks <sup>[27]</sup>. In the case of bottle-fed infants, drinking-water can be the major external source of exposure to nitrate and nitrite. A guideline value of 50 mg/L for nitrate ion is established by WHO to protect against methaemoglobinaemia in bottle-fed infants. Absorption of nitrate ingested from vegetables, meat or water is rapid and in excess of 90 %, and final excretion is in the urine. Contaminated private wells have been associated with most cases of methaemoglobinaemia. In the present study, nitrate ion concentration in the water samples range between 3.28–12.27 mg/L. This values satisfy the WHO recommendation of maximum of 50 mg/L for nitrate ion. Natural sources and industrial waste water are the main sources of sulphate in groundwater. The existing data do not identify a level of sulphate in drinking-water that is likely to cause adverse human health effects <sup>[27]</sup>. Therefore, healthbased guideline value has not been derived for sulphate. However, a laxative effect may occur at concentrations of 1000–1200 mg/L. The presence of sulphate in drinking-water may also cause noticeable taste and may contribute to the corrosion of distribution systems. Taste thresholds have been found to range from 250 mg/L for sodium sulphate to 1000 mg/L for calcium sulphate ion. It is generally considered that taste impairment is minimal at levels below 250 mg/L of sulphate. In the study area, water samples collected have sulphate concentration, which ranges from 14.43–31.19 mg/L and thus satisfy the WHO recommended maximum value. Based on the results from the geophysical, geotechnical and groundwater quality assessment, suitable and non-suitable areas for the proposed sanitary landfill have been unravel (Figure 4).





# 5. Conclusions

The suitability of the soil as landfill liners have been evaluated based on geophysical and geotechnical parameters. Most of studied soils satisfy the recommended values of each parameter and are considered suitable for use as landfill liner. The results of the geophysical and geotechnical assessments indicate that the soils in the northern part of the area satisfy the recommended values of each parameter and are considered suitable for siting the sanitary landfill. In addition, this soil is preferred to their southern counter part due to the presence of thick layer of clay as well as the absence of fractures and shear zones which may increase the risk of groundwater pollution. Geotechnical analysis also revealed that the soil samples obtained from the northern part of the area are less permeable, thus they can act as seals in sanitary landfill. The generated suitability map indicate that approximately 45% of the landmass is suitable for sanitary landfill construction. Assessment of baseline groundwater quality shows that physicochemical parameter of the water samples are generally below the values recommended by World Health Organization (WHO). However, the high concentration of Nickel (Ni<sup>2+</sup>), Lead (Pb<sup>2+</sup>) and Arsenic (As<sup>3+</sup>) in most of the water samples point to the fact that present dumping activities on the site has negatively impacted the groundwater quality in the

area. Hence, the consumption of the water over a long period of time would be harmful to both humans' health and animals.

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To whom correspondence should be addressed: Professor Olusegun Omoniyi Ige, Department of Geology and Mineral Sciences, University of Ilorin, Private Mail Bag 1515, Ilorin, Kwara State, Nigeria, *E-mail:vickyige2002@yahoo.com; olusegun@unilorin.edu.ng* 

# Article

The Effect of Leachate Contaminant on the Geotechnical Properties of Lateritic Soils: A case study of lateritic soils around Ilorin in Kwara State, Nigeria

Olusegun Omoniyi Ige<sup>1</sup> and Christopher Baiyegunhi<sup>2</sup>

- <sup>1</sup> Department of Geology and Mineral Sciences, University of Ilorin, Private Mail Bag 1515, Ilorin, Kwara State, Nigeria
- <sup>2</sup> Department of Geology and Mining, School of Physical and Mineral Sciences, University of Limpopo, Private Bag X1106, Sovenga 0727, Limpopo Province, South Africa

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#### Abstract

The way out of hitches in soil engineering requires a detailed knowledge of the geotechnical properties of soils, which are, perhaps, one of the most complex materials to be studied. This is partly because geotechnical properties of soils are affected by several factors including leachate-contaminant. The aim of the study is to determine the effects of the leachate-contaminant on soils' behavior when used for engineering purposes. In order to determine the effects of leachate contamination on geotechnical properties of lateritic soils, four portions of lateritic soil samples collected around Ilorin and artificially contaminated with 0%, 5%, 10% and 20% leachate and their respective geotechnical properties such as specific gravity, particle size distributions, consistency limits and compaction were determined. The geotechnical results show that leachate contamination proportionally increased with the Atterberg limits and permeability values for the contaminated soils, but decreases in compaction parameters. For compaction tests, maximum dry density decreased from 15.9 kN/m<sup>3</sup> in the control sample to 14.4 kN/m<sup>3</sup> in contaminated soil at 20 % leachate concentration. The co-efficient of permeability values for soils contaminated with 0 %, 5 %, 10 % and 20 % leachate are  $1.6 \times 10^{-6}$  cm/s,  $1.8 \times 10^{-6}$  cm/s,  $2.3 \times 10^{-6}$  cm/s and  $2.8 \times 10^{-6}$  cm/s, respectively. The coefficient of permeability values increased with increase in leachate content. It was observed that the addition of leachate has adverse reducing effects on the strength and hydraulic properties of the contaminated residual soil. The study revealed that the use of leachate-contaminated soils in geotechnical engineering works should be avoided because it is inimical to life, money and properties.

Keywords: Leachate; Contaminated soil; Residual soil; Atterberg limits; Geotechnical properties.

## 1. Introduction

Human population is increasing on daily basis, so is the corresponding quantity of waste contending for space with man and its effect impairing the quality of environment <sup>[1]</sup>. In recent time, the impact of leachate or impurities on groundwater and soil have attracted a lot of attention because of its overwhelming environmental significance. The leachate from irregular landfills links directly to groundwater, and pollutes the groundwater as well as the hosting or local soil. Lateritic soils constitute an important unit of soil in parts of Nigeria <sup>[2-3]</sup>. It is common and abundantly found as near surface material (below humus layer) within soil profile, especially in the southwestern Nigeria <sup>[4]</sup>. Lateritic soil, in its natural existence sometimes possess good engineering properties that have made it useful for engineering works like roads, dam, building bricks and barriers in sanitary landfills <sup>[5-11]</sup>. Lately, due to increased urbanization and waste generation in Nigerian cities, this unit of soil layer has been turned to repository of solid and liquid waste by government agencies and individual. Leachate generation within the waste is encouraged by the process of the decay of organic component of the waste in the

presence of chemically active water. According to Stevenson and Buttler <sup>[12]</sup>, humic substances with functional group such as carboxyl, carbonyl and phenolic hydroxyl are produced from the decay of organic component.

The effects of different impurities on natural properties of lateritic soils have been studied by several researchers with various deductions <sup>[1, 13-14]</sup>. Sunil et al. <sup>[15]</sup> reported that the chemical and geotechnical properties of lateritic soils are altered by increasing content of pH, while Indrawan et al.<sup>[16]</sup> documented that increasing amount of coarse-grained materials increased the saturated permeability and reduced the shrinkage potential of residual soil. A number of other related studies were carried out in order to investigate the geotechnical behavior of oil contaminated soil <sup>[17]</sup>. Most of the report shows that oil contamination significantly reduced Atterberg limits, maximum dry density, permeability and strength properties of soils, which in turns affects their use for any engineering construction work. George and Beena <sup>[18]</sup> artificially contaminated soil with proportion of municipally generated leachate and discovered that permeability and shear strength increased, while consistency limits values reduced with increasing percentage of leachate. In cities of Nigeria, sources of wastes generation are on the increase and irrational disposal of these wastes on open or excavated lands/dumpsite creates source of soil pollution due to generation of leachate. These dumpsites, when closed or abandoned are commonly acquired by government agencies or individual for development of civil engineering works such as shopping complex or mall, bank and residential houses etc. The effects of leachate-soil interaction on the underlying soil at such sites in Nigeria have not been well investigated or documented. However, such interaction may weaken the strength of soil <sup>[19]</sup>, thus limiting their application in civil engineering works or making the superstructure a potential death trap. Pursuant to the above, this study was undertaken to investigate the effects of leachate contamination on some geotechnical properties of lateritic soil around Ilorin in Kwara State, Nigeria (Figure 1).



Figure 1. Map showing the study area and sampling site (modified after Ige et al. [8])

Geologically, the study area lies in the Precambrian Basement Complex area of southwestern Nigeria and is underlain by rock of metamorphic and igneous types <sup>[5]</sup>. The hydrologic setting of the area is typical of what is obtained in other Basement complex area; where the availability of water is a function of the presence of thick little clay overburden material and presence of water filled joints, fracture or faults within the fresh Basement rocks.

## 2. Materials and methods

The study was conducted within 1 year, between September 2020 and March 2021. About 5 kg of lateritic sample, derived from a granite-gneiss along Asa dam road, 500 m to Dangote Flour Mill (Figure 1) was collected for the purpose of preparing the leachate-contaminated samples. In order to ensure uniform conditions, sampling was limited to an area of one square meter within the same horizon. The samples were collected with the aid of digger, shovel and clean polythene bags at a depth of about 4m. At this depth, the sample is free from influence of plant roots, which may affect its properties. The sample was collected and stored in an air-tight plastic container before being air dried at room temperature for 72 hours. The sample was pulverized into powdery state and air-dried for two weeks.

To prepare the leachate-contaminated soil, 10 liters of raw sample of leachate was collected from the base of over 20 years old waste dumpsite (at Amilegbe Bridge) in Ilorin, Nigeria. The dumpsite receives both hazardous and non-hazardous waste (domestic, industrial, hospital etc), which are the common waste generated in this part of the country <sup>[14]</sup>. The temperature, density and colour of the leachate are 21°C, 0.786 kg/l and darkish brown, respectively. A filter paper was placed on each face of the soil specimen to prevent the clogging of the perforated disks by the soil fines. After placing the bottom and top plate of the falling head permeameter, the nuts were fastened and assembled properly. The permeameter was then connected to a standpipe (when testing uncontaminated, soil the standpipe was filled with distilled water and during testing of contaminated soil, the standpipe was filled with leachate). The soil was saturated by allowing permeant (leachate) to flow continuously through the sample from the standpipe. Saturation of the soil sample was ensured under steady state flow conditions. The coefficient of permeability was calculated with the equation below:

$$k = \frac{2.303aL\left(\log\left[\frac{h_1}{h_2}\right]\right)}{At}$$

where k = coefficient of permeability; a = cross sectional area of stand pipe (cm<sup>2</sup>); L = height of soil sample (cm); A = cross sectional area of sample (cm<sup>2</sup>);  $h_1$  and  $h_2 = \text{initial and final height of permeant in stand pipe (cm) and t = time taken for drop from <math>h_1$  to  $h_2$  (sec).

The contaminated soil was divided into three parts, with each containing different proportions of leachate in order of 5%, 10% and 20% of dried weight. The contaminant was thoroughly mixed with soil and the mixture was permitted to cure in closed container at ambient temperature for 7 days. One uncontaminated sample was used as a control sample to monitor and establish the relationships between leachate contaminants and geotechnical properties of soils. Thereafter, preliminary geotechnical classification and identification tests such as particle size distribution analyses. Particle size distribution, specific gravity, dry and bulk densities, Atterberg consistency limits, compaction and coefficient of permeability tests were carried out on the four soil samples based on the British Standard (BSI 1377:1990). Each geotechnical test was performed twice on the same soil sample under the same condition in order to determine the reliability of the geotechnical test results.

# 3. Results

The basic geotechnical properties of the soil samples are presented in Table 1 and Figures 2-4. The grain size distributions result revealed 6% gravel, 47% sand and 47% fine (Table 1). The liquid limit, plastic limit and plasticity index values for uncontaminated soil with 0% leachate are 48.5%, 23.3% and 25.2%, respectively. The liquid limit, plastic limit and plasticity index values for the soil contaminated with 5% leachate are 51.4%, 25.2%, and 26.2%, respectively. The liquid limit, plastic limit, plastic limit and plasticity index values for the soil contaminated with 5% leachate are 51.4%, 25.2%, and 26.2%, respectively. The liquid limit, plastic limit, plastic limit and plasticity index values for the soil contaminated with 5% leachate are 51.4%, 25.2%, and 26.2%, respectively. The liquid limit, plastic limit and plasticity index values for the soil contaminated with 5% leachate are 51.4%, 25.2%, and 26.2%, respectively. The liquid limit, plastic limit and plasticity index values for the soil contaminated with 5% leachate are 51.4%, 25.2%, and 26.2%, respectively. The liquid limit, plastic limit and plasticity index values for the soil contaminated with 5% leachate are 51.4%, 25.2%, and 26.2%, respectively. The liquid limit, plastic limit and plasticity index values for the soil contaminated with 5% leachate are 51.4%, 25.2%, and 26.2%, respectively.

with 10 % leachate are 56.3%, 28.4% and 27.9%, respectively. The liquid limit, plastic limit and plasticity index values for the soil contaminated with 20 % leachate are 61.7%, 33.4% and 28.3%, respectively. The specific gravity values increased from a maximum of 2.63 in sample contaminated with 20% leachate up to 2.67 in uncontaminated sample. On the contrary, the bulk density values ranged a minimum of 1.27 g/cm<sup>3</sup> in uncontaminated sample to a maximum of 1.83 g/cm<sup>3</sup> in sample contaminated with 20% leachate. For uncontaminated soil, the Optimum Moisture Content (OMC) and Maximum Dry Density (MDD) are 20.3% and 15.9 KN/m<sup>2</sup>, respectively; while the 5% leachate contaminated soil has OMC and MDD values of 20.8% and 15.6 KN/m<sup>2</sup>, respectively (Figure 2). The OMC and MDD values for the 10% leachate contaminated soil are 22.0% and 15.5 KN/m<sup>2</sup>, respectively, and 24.5% and 14.4 KN/m<sup>2</sup>, respectively for the 20% leachate contaminated soil. The co-efficient of permeability values for soils contaminated with 0 %, 5 %, 10 % and 20 % leachate are  $1.6 \times 10^{-6}$  cm/s,  $1.8 \times 10^{-6}$  cm/s,  $2.3 \times 10^{-6}$  cm/s and  $2.8 \times 10^{-6}$  cm/s, respectively (Figure 4).

Parameters	Control sam- ple	Percentage	ercentage of dry weight of base oil				
	0.0 %	5.0%	10.0%	20.0%			
Atterberg limits							
Liquid limit	48.5	51.4	56.3	61.7			
Plastic limit	23.3	25.2	28.4	33.4			
Plasticity index	25.2	26.2	27.9	28.3			
Specific Gravity	2.67	2.67	2.66	2.63			
Density tests (g/cm <sup>3</sup> )							
Bulk density	1.27	1.72	1.79	1.83			
Dry density	1.25	1.18	1.18	1.20			
Compaction tests							
Optimum moisture content (%)	20.3	20.8	22.0	24.5			
Maximum dry density KN/m <sup>2</sup> )	15.9	15.6	15.5	14.4			
Grain size distribution of control sample	Gravel=6%	Sand = 47%,		Fine =47%			

Table 1. Geotechnical characteristics of control and oil-contaminated soil samples.



Figure 2. Compaction curves for lateritic soils with different leachate content



Figure 3. Relationship between compaction parameters and leachate content



Figure 4. Variation in coefficient of permeability at different leachate content.

#### 4. Discussion

The summary of results obtained from moisture content, liquid limit, plastic limit and plasticity index analyses are presented in Table 1. It is clear from that liquid limit, plastic limit and plasticity index increase with increase in leachate content. This trend may be due to polarized nature of the moisture content. The polarized water is attracted by negatively charged clay surface, thus influences the orientation of water around clay particles. Gillot <sup>[20]</sup> reported that clay soils with non-polarized fluid do not have plasticity properties. Crude oil-waste is a nonpolarized fluid, as the oil is evenly mixed with the samples; it covers the grains and reduces water-soil interaction. This brings about increase in the consistency limits as the leachate content increases. The compaction is carried out to improve qualities of soils for suitability in engineering construction works. Light energy (standard proctor) of compaction was used to compact both control and artificially contaminated samples. By comparing the values of the optimum moisture content (OMC) and the maximum dry density (MDD) in the uncontaminated sample and contaminated samples, there was a noticeable departure in the values of OMC and MDD of uncontaminated and contaminated soils as the leachate content increases. OMC increases with increasing content of leachate, while MDD decreases as the leachate content increases. This result is consistent with other related studies by Shah et al. [21] on oil contaminated soils. The MDD values for contaminated soils showed a sharp and consistent departure from 15.9kN/m<sup>2</sup> obtained for control samples to 15.6-14.4kN/m<sup>2</sup> in contaminated soil as the proportion of leachate increases. These results are consistent with Shah et al. <sup>[21]</sup> study on oil-contaminated material. For the energy of compaction, the MDD consistently decrease with increase in leachate content while the OMC increases (Figure 3). These could be due to the lubricating effect of the presence of leachate, which prevents effective compaction. Also, the presence of leachate reduces the amount of water content needed to reach MDD. The results are similar to findings of Al-sanad et al. <sup>[3]</sup> who reported that at excess oil-waste content, the shape of the curve will be odd. The coefficient of permeability of any soil is dependent on several factors such as fluid velocity, pore-size distribution, grain size distribution, void ratio, roughness of mineral particles, and degree of saturation <sup>[22]</sup>. With the falling head permeability tests carried out on both contaminated and uncontaminated samples to compare the coefficient of permeability, there was increase in leachate concentration as the co-efficient of permeability of the contaminated soil increases (Figure 4). This increase in coefficient of permeability is attributed to chemical reaction between the leachate and the clay minerals. It is reported that strongly acidic and strongly basic liquids can dissolve clay minerals <sup>[23]</sup>. The dissolution of clay mineral particles by leachate increases the effective pore space and hence, the hydraulic conductivity increases. Sunil *et al.* <sup>[15]</sup> reported that the cementing agents in soils help to bind the finer particles together to form aggregates. However, strongly acidic conditions lead to the destruction of soil structure. Hence, as the particles are percolated by permeant, they clog pore spaces. Nevertheless, as dissolution progresses in the zones of clogging, particles will be removed and the hydraulic conductivity increases.

#### 5. Conclusions

The effects of leachate contamination on some geotechnical properties have been investigated on lateritic soils with the index, compaction and permeability test performed in accordance with the BS standard. The Atterberg limits values of contaminated soils were higher than that of uncontaminated soil, while the maximum dry densities also dropped as leachate content increases above 5% in contaminated soils. Similar behavior was observed on compaction and co-efficient of permeability properties of the investigated soils. There was a noticeable departure in the values of OMC and MDD of control and contaminated soils as the leachate content increases. OMC increases with increasing content of leachate, while MDD decreases. The co-efficient of permeability values also increase in values with increasing percentage of leachate content in contaminated soils. The results showed that leachate contamination on soil has adverse influence on geotechnical properties of lateritic soil. Thus, use of old or abandoned open dumpsites civil engineering construction of superstructures should be discouraged and discontinued because of potential defects on soil strength, toxic consequences on lives, properties and eventual wastage of money.

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#### Competing interest

The authors have declared that no competing interest exists.

#### Data availability

All relevant data are within the paper and its supporting information files.

#### Significance statement

This study discovered that as the percentage or concentration of leachate contaminants increases in lateritic soil, the bulk density, Atterberg limits, permeability and OMC values increases, while there is decrease in MDD values, hence, reducing the strength and hydraulic properties of the soil which in turns affects their use for engineering construction work. This study can be beneficial government agencies or individual because it will serve as a guide and inform them to check leachate-contaminant in laterite soil before they use it for using for any engineering constructions works. Thus, it will in turn help in avoiding the potential death trap and losses that such soil possess.

#### Authors' contribution

*Olusegun Omoniyi Ige conceived and supervised the work. Christopher Baiyegunhi wrote and revised the paper.* 

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To whom correspondence should be addressed: Professor Olusegun Omoniyi Ige, Department of Geology and Mineral Sciences, University of Ilorin, Private Mail Bag 1515, Ilorin, Kwara State, Nigeria, E-mail: vickyige2002@yahoo.com; olusegun@unilorin.edu.ng

# Article

# **Open Access**

Geotechnical and Mineralogical Evaluation of Lateritic Soils to Access Their Suitability for Engineering Construction Works: A Case Study of Some Selected Residual Soils in Parts of the Southwestern Nigeria

Morakinyo Ebunoluwa<sup>1</sup>, Olufemi Ogunsanwo<sup>1</sup>, Olusegun Omoniyi Ige<sup>1</sup>, and Christopher Baiyegunhi<sup>2</sup>

- <sup>1</sup> Department of Geology and Mineral Sciences, University of Ilorin, Private Mail Bag 1515, Ilorin, Kwara State, Nigeria
- <sup>2</sup> Department of Geology and Mining, School of Physical and Mineral Sciences, University of Limpopo, Private Bag X1106, Sovenga 0727, Limpopo Province, South Africa

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#### Abstract

This study investigate the geotechnical and mineralogical properties of differently sourced lateritic soils in order to determine their suitability for engineering construction purposes. In addition, attempts were made to establish the relationships between mineralogy and some geotechnical/engineering properties. Five lateritic soil samples derived from quartzite, mica schist, amphibolite, sandstone and migmatite-gneiss in parts of the southwestern Nigeria were collected in the field for geotechnical tests as well as determine their mineralogical compositions using x-ray diffraction (XRD). Result from grain size analysis shows that the soils are generally fine to medium grained consisting mainly of clayey silty sand. The Atterberg consistency limits revealed that the soils contain inorganic clay of low to intermediate plasticity, with linear shrinkage limit between 7% and 10%, indicating medium shrink potential. Based on the Skempton's activity chart, the soil can be classified as active to moderately active soils. The maximum dry density values of the samples at standard proctor energy of compaction range from about 1.46 g/cm<sup>3</sup> to 1.96 g/cm<sup>3</sup>, while those obtained at modified proctor energy vary between 1.57 g/cm<sup>3</sup> and 2.09 g/cm<sup>3</sup>. The shear strength tests give angle of internal friction between 31° and 33° and cohesion values ranging between 59kPa and 70kPa for the standard and modified proctor compaction, respectively. These values indicate that the soils will have high bearing capacity and could support a moderately steep slope. The coefficient of permeability of the soils is low with kranging from 0.7 x 10<sup>-8</sup> m/sec to 8.79 x 10<sup>-8</sup> m/s, which falls within the range considered to be impermeable. The soaked and unsoaked California Bearing Ratio (CBR) values vary from about 1% up to 6%. Based on the geotechnical analysis, the soils can be used as liners in landfill system, fill materials in embankments and earthfill dam construction as well as subgrade and fill materials for road pavement in highway construction. The mineralogical analysis showed that the soils are mostly composed of illite, kaolinite, guartz, feldspar, hematite and goethite.

Keywords: Geotechnical; Mineralogical; Lateritic soils; Engineering; Nigeria.

## 1. Introduction

Civil engineering construction works in Nigeria are beset with multi-facetted problems <sup>[1-4]</sup>. Devastating flood from excessive rainfall are accelerating the failure process, which results in immense damage to agriculture and infrastructures every year <sup>[5]</sup>. Over the last few decades, there have been several cases of structures failure that were initially thought to be well engineered. The major causes of these failures were identified to be erosion, seepage and sliding. Furthermore, insufficient supervision during construction results in poor-quality earthworks with the use of inappropriate soil materials, insufficient or no clod breaking, inadequate compaction and the use of inferior materials as well as inadequate maintenance <sup>[6-7]</sup>. Among many reasons, the improper design, methodology and construction procedure is prime and one of

the most important causes of failure in engineering structures <sup>[8]</sup>. However, clear understanding of material behavior is necessary to interpret the failure phenomenon of a particular problem.

The response of Engineering geologists to the growth in developmental projects, the difficulty in understanding soil conditions and the failures associated with tropical soils, and the need to address these failures, has led to the apparent increase in research on the tropical soil types and their engineering properties, especially laterite soils <sup>[9-10]</sup>. The importance of these laterite soils cannot be more emphasized as they are being used as construction and engineering material for roads and airfield sub-bases and sub-grades; fills and embankments for bridges and dams and other engineering uses as may require soil materials in the tropics [11-14]. Several authors like Baiyegunhi et al. <sup>[4]</sup>, Gidigasu <sup>[10]</sup>, Ige <sup>[11]</sup>, Jegede <sup>[14]</sup>, and Ogunsanwo <sup>[14-15]</sup> have worked on the geotechnical properties of lateritic soils, since their properties change even within few meters, and have reported topography, climate, soil type, parent material etc., as major controlling factors of their engineering properties. Suitability of lateritic soils as base and subbase course material in road construction and barriers in sanitary landfill have been reported with no particular interest in the influence of rock types <sup>[4,11-17]</sup>. Thus, this study focuses at comparing the geotechnical properties in relation to the mineralogy of soils derived from five different rock types. Rocks of the study area can be broadly divided into the Basement Complex rocks and sedimentary rocks (Figure 1). Four of the lateritic soils under consideration are derived from Basement Complex rocks. The fifth sample is derived from the Santorian-Maastrichian units (Nupe sandstone) of the Lower Bida Basin.





## 2. Materials and methods

Field investigation was carried out within the study area to collect soil samples. A total of five samples with different parent rock were obtained in road-cut exposures of laterites soils in Omu-Aran, Osho, Ife-Ifewara, Share and Oko (Figure 1). The five lateritic soil samples, labelled QTZ, MCS, AMP, NPS and MIG were collected from the aforementioned locations are derived from quartzite, mica schist, amphibolite, sandstone and migmatite-gneiss, respectively. These rock types are among the most widespread rock types in these areas. The collected fresh soil samples were kept in a neat and labelled polythene bag. The samples were sun dried for 10 days to ensure removal of moisture content. The index properties of the soil including specific gravity, moisture content, linear shrinkage, bulk density and dry density determination, Atterberg limits and grain size analysis were determined in accordance with British Standard (BS 1377) test procedures. Other test carried out are dispersion test, swelling test, permeability test (falling head), standard and modified proctor compaction, California

bearing ratio (CBR), direct shear test and mineralogy. Each geotechnical test was performed at least twice on the same soil sample under the same condition in order to determine the reliability of the geotechnical test results.

#### 3. Results and discussion

## 3.1. Specific gravity

The summary of geotechnical tests results is presented in Table 1. The specific gravity of the soils range from 2.55 – 2.70. The obtained specific gravity values are indicative of a high degree of laterization of the soils <sup>[5]</sup>. Tuncer and Lohnes <sup>[8]</sup> noted that specific gravity is important in estimating the degree of weathering of soils. Rocks rich in heavy minerals weather to soils whose grains have high values of specific gravity. Also, soils that are rich in iron and titanium bearing compounds have high values of specific gravity. According to Ramamurty and Sitharam <sup>[20]</sup>, the studied soils can be classified as inorganic soils based on the specific gravity. Amphibolite derived soil (AMP) has the highest value of 2.70, while Nupe Sandstone (NPS) derived soil has the least value of 2.55. The results place, in order of competence, solis derived from amphibolite (AMP), mignatitte (MIG) and quartzite (QTZ) as the more suitable in road and building construction when compare to other soil analyzed.

Traverse	VES sta- tion	No of layers	Apparent resistivity (Ωm)	Thickness (m)	Depth to base- ment (m)
	1	4	796/1883/903/4455	1.45/8.38/17.4/	27.2
	2	4	120/522/685/1000	1.18/6.57/16.6/	24.4
1	3	4	74.2/415/755/2663	1.19/4.92/15.9/	22.0
	4	4	742/1474/590/3026	1.28/5.17/13.4/	19.8
	5	4	916/3871/1579/4780	1.47/11.8/8.38/	21.6
	6	4	613/3190/1018/4249	1.0/6.99/15.7/	23.7
_	7	4	461/850/554/5652	1.88/7.72/11.7/	21.3
2	8	4	231/1503/554/4080	1.13/8.09/12.0/	21.2
	9	4	490/1699/922/6790	1.13/2.79/15.3/	19.2
	10	3	5426/2040/8849	2.31/15.2/	17.5
3	11	3	356/478/1000	0.5/6.05/	6.6
	12	3	686/416/1570	2.17/4.76/	6.9
	13	4	2945/408/1818/4455	1.08/4.92/8.72	14.7
4	14	3	1519/3334/7682	2.1/3.49/	5.6
	15	3	2659/1279/7483	1.02/3.51/	4.5
	16	3	752/2353/5599	2.17/3.94	6.1
	17	3	3799/1264/6453	0.79/6.58	7.4
5	18	3	1000/2259/5427	1.18/4.08	5.3
	19	3	110/736/5934	0.13/7.0/	7.1

Table 1. Summary of geotechnical tests results of soil samples

## 3.2. Grain size distribution

The grain size analysis was performed to determine the particle size distributions in the soil. The distribution of different grain sizes affects engineering properties of soil, which in turn influences their usability in construction works. The summary of grain sizes distribution parameters of the soil samples are shown in Table 2. It is evident from the Table 2 and Figure 2, sand and silt fractions are predominant in the particle size distribution of the soils. The mica schist derived soil (MCS) has the highest amount of fines (38%), while the amphibolite derived soil (AMP) has the lowest amount of fines (30%). The plot of the soils in the AASHTO classification system shows that the quartz schist (QTZ) and amphibolite derived soils (AMP) plotted in the A-2-6 field, Sandstone derived (NPS) and mica schist derived (MCS) soil falls under A-6, whereas migmatite gneiss derived soils (MIG) falls under A-2-7. All the soils are grouped as clayey silty sand based on the grain size fractions. FMWH <sup>[9]</sup> stipulated amount of fines is  $\leq$ 

35% for use as sub-base materials. On the basis of this, the soils except MCS fall into the category and therefore they have good potential in the construction industry as they will be suitable for use as road sub-base material. Furthermore, various authors have affirmed that lateritic soil might be useful as landfill liners if the fine component is greater than 30% <sup>[13,21-30]</sup>. Thus, all the studied soils are useable as landfill in sanitary.

	WC	BD				Compactio	on Test	Permeability.
Sample	(%)	(kg/m <sup>3</sup> )	SG	USC	Atterberg Limit	Standard	Modified	k (m/sec)
T1	1.3	1.28	2.55	SC- SM	LL= 39%; PL= 21%; Ip = 21%; FI= ; TI= AC=0.33 ; SL=	OMC = 16 %; MDD =1.8g/cm <sup>3</sup>	OMC = 14 %; MDD =1.83g/cm <sup>3</sup>	4.892 x 10 <sup>-7</sup>
Т2	2.8	1.26	2.64	SC- SM	LL= 35%; PL= 21%; Ip =14% ; FI=22 ; TI=0.64; AC=0.28 ; SL=	OMC =16.5 %; MDD =1.73g/cm <sup>3</sup>	OMC = 16 %; MDD =1.78g/cm <sup>3</sup>	4.467 x 10 <sup>-7</sup>
Т3	0.6	1.66	2.60	SP		OMC = 10 %; MDD =1.8g/cm <sup>3</sup>	OMC = 9 %; MDD =1.83g/cm <sup>3</sup>	1.361 x 10 <sup>-6</sup>
T4	3.7	1.38	2.66	SC- SM	LL=30% ; PL=18% ; Ip =21% ; FI= ; TI= AC= 0.33; SL=	OMC = 15.5 %; MDD =1.8g/cm <sup>3</sup>	OMC = 14.5%; MDD =1.85g/cm <sup>3</sup>	6.080 x 10 <sup>-7</sup>
Т5	2.1	1.65	2.65	SC- SM	LL=30% ; PL=17%; Ip =13% ; FI= 25; TI= 0.5; AC=0.6 ; SL=	OMC =12 %; MDD =1.79g/cm <sup>3</sup>	OMC = 11 %; MDD =1.82g/cm <sup>3</sup>	1.101 x 10 <sup>-6</sup>
Т6	5.9	1.35	2.60	SC- SM	LL= 34%; PL=23%; Ip =11%; FI= 25; TI= 0.6; AC=0.2; SL=	OMC = 16 %; MDD =1.7g/cm <sup>3</sup>	OMC = 14 %; MDD =1.76g/cm <sup>3</sup>	5.142 x 10 <sup>-7</sup>
BH1	14.6	1.82	2.55	SC	LL= 24%; PL=16% ; Ip = 7%; FI= ; TI= AC= ; SL=	OMC = ; MDD =	OMC = ; MDD =	
BH2	11	1.85	2.64	SC	LL=28% ; PL=17% ; Ip = 11%; FI= ; TI= AC= ; SL=	OMC = ; MDD =	OMC = ; MDD =	

Table 2. Summary of the distribution of grains within soil samples





## 3.3. Atterberg consistency limits

The results of the Atterberg consistency limits and classification tests is presented in Table 1. The soils have liquid limit ranging between 29% - 41% and plasticity index between 13.8% -19.6%. According to FMWH <sup>[9]</sup> specification, subgrade materials should have a liquid limit  $\leq$ 50% and plasticity index  $\leq$  30%, while for subbase, liquid limit should be  $\leq$  30% and plasticity index  $\leq$  12%. According to Wright <sup>[30]</sup>, the liquid limit values of 40% and above are assumed high in pavement construction. Furthermore, they reported that plasticity index value of 10%and above are also assumed high in pavement design. All the soils meet the requirement for use as subgrade materials since they have plasticity index values within the threshold. Atterberg limits are also important in the selection of materials for use as liners in landfill systems. Benson *et al.* <sup>[6]</sup> recommended that the liquid limit of the liner materials must be at least 20%. Most of the specification for soil liners proposed by various researchers or waste regulatory agencies does not generally prescribe any limit (maximum value) for the liquid limit [10-12]. As long as it does not create any working problem, soils with high liquid limit are generally preferable because of their low hydraulic conductivity <sup>[10,21]</sup>. The liquid limit of all the studied soil is higher than the minimum prescribed value. The value also falls within the range obtained by Iqe [11,13], hence could be promising for use as barriers in landfill systems.

## 3.4. Clay activity (Ac)

The 'activity' of the soil samples can be calculated from the relationship between the plasticity index and percentage of clay in a particular sample by weight.

Ac = PI/ (% of clay-sized fraction by weight)

where A = Activity, PL = Plastic Index

The calculated `activity' of the soils is shown in Table 1. The plastic properties of the soils result from the adsorbed water that surrounds the clay particles. This is directly related to the type of clay minerals and their amount in the soil, which in turn affect the liquid limit and plastic limit <sup>[21-23]</sup>. The clay activity results revealed that the soils exhibit activity Casagrande's plot which shows that the samples can be interpreted as inorganic clay of low plasticity (Figure 3), ranging from 0.5-2.1 and 0.7-2.2 for unstable and stable locations, respectively.



Figure 3. Positions of the soil samples in the Casagrande's plasticity chart

# 3.5. Free swell

Swelling soils are often deleterious when used directly in constructions works. This is because of the ability of these soils to absorb high moisture. Both the amount of swelling and magnitude of swelling pressure are known to be dependent on the clay minerals, the soil mineralogy and structure, fabrics and several physic-chemical aspects of the soil <sup>[24-26]</sup>. Based on the free swell test result shown in Table 1, the studied soil samples with the exception of sample MCS, tend to be non-expansive. The MCS soil had intermediate degree of expansive-ness, which is probably due to the occurrence of Illite as one of its major minerals. Hence, it has to be subjected to some form of treatment before it can be utilized successfully in construction works.

## **3.6. Dispersive test**

Soil gets dispersed and washed away in water. This process is termed as dispersiveness. Non-plastic nature of the soil particles and its inadequate inter-particles attraction causes dispersal. Dispersive property of soil cannot be identified by the standard laboratory index tests, such as visual classification, grain size analysis, specific gravity or Atterberg limits. In this study, crumb test, double hydrometer test and turbidity tests have been performed to identify dispersive clays. It is observed that MCS is highly dispersive, while MIG, NPS, QTZ and AMP tend to have intermediate-high dispersiveness. The presence of dispersive soils in the embankment dam or foundation has been said to be one of the factors contributing to failures in structures which are reasonably well engineered <sup>[2,16-17,26]</sup>.

# 3.7. Compaction test

The compaction test result is presented in Table 1. It can be seen that the optimum moisture content (OMC) of the soils showed a decrease at the modified proctor energy when compared to the value at the standard proctor energy. At standard proctor compaction, the QTZ, MCS and MCS samples have the highest MDD of 1.81g/cm<sup>3</sup>, 1.70g/cm<sup>3</sup> and 1.63g/cm<sup>3</sup>, respectively. The AMP and MIG lateritic soils have the lowest, with equal MDD value of 1.57g/cm<sup>3</sup>. At the energy of the modified proctor, the QTZ, MCS and NPS lateritic soils have the highest MDD of 1.85g/cm<sup>3</sup>, 1.80g/cm<sup>3</sup> and 1.75g/cm<sup>3</sup>, respectively. The AMP and MIG lateritic soils have the lowest MDD values of 1.65g/cm<sup>3</sup> and 1.64g/cm<sup>3</sup>, respectively. The MIG lateritic soil has the highest moisture content of 18% at standard proctor energy, it also has the highest moisture content of 17% at modified proctor energy. The OMC serves as a guide to know the amount of water required during construction. Thus, MIG requires the highest water content on the field to achieve maximum dry density at low energy of compaction. However, the values generally fall within the values that were recommended by Baiyegunhi *et al.* <sup>[4]</sup>, Ige <sup>[11,13]</sup>, Ogunsanwo <sup>[14-15]</sup> and Jegede <sup>[17]</sup>, for materials before they can be used fills in dams, building, and base course in road and liner in landfill.

# 3.8. Permeability test

The result of the co-efficient of permeability of the remolded soil samples is depicted in Table 1. The values of permeability co-efficient range from  $10^{-8}$  to  $10^{-11}$  m/s, which falls into the soils classified as impervious soils <sup>[21,27]</sup>. It is considered that such practically impermeable soils will be useful as fill materials in values much lower than 7.54x10<sup>-10</sup> and 1.12 x 10<sup>-11</sup> m/sec. Thus, this make the MSC and AMP samples most suitable as barrier in sanitary landfill dam constructions. Several researchers have also suggested a maximum coefficient of permeability value  $1x10^{-9}$ m/sec for landfill barrier soils <sup>[7,19,28-30]</sup>.

# 3.9. California bearing ratio (CBR)

The Nigerian specification for roads and bridges as stipulated by FMWH <sup>[8]</sup> is a minimum CBR of 8% for sub-grade/fill, while the asphalt <sup>[1]</sup> stipulated CBR value of between 0% to 3% for sub-grade and 3% to 7% for sub-base. In align with the aforementioned values, the studied lateritic soils can be classified as having poor to fair CBR and can only be used as subgrade and fill materials in highway construction. Highest CBR value was recorded in QTZ under unsoaked condition through modified proctor energy, while the least CBR value was recorded in MSC under soaked condition through standard proctor energy (Table 1). This may be connected to the ability of the clay mineral to absorb water. The MSC sample has illite as major mineral, which is responsible for it swelling and shrinking attribute.

# 3.10. Shear strength

The computed values of the undrained shear strength parameters, C and  $\phi$  are shown in Table 1. For the shear tests carried out using samples compacted at standard proctor, the NPS lateritic soil has the highest cohesion value (45 kPa), while the AMP lateritic soil have the lowest cohesion value (25 kPa) at this same energy. At modified Proctor energy, the QTZ soil has the highest cohesion value (120 kPa), while the cohesion value of 30 kPa, which is the lowest was recorded for MCS, AMP and MIG lateritic soils. For lateritic residual soils, Mitchell and Sitar <sup>[21]</sup> reported that for compacted samples the majority of the friction angles range ( $\phi$ ) from 28° to 38° and cohesion values range (C) from 0 to 48 kPa. The results obtained in this study is close or similar to the values recommended by Mitchell and Sitar <sup>[16]</sup> for cohesion (C) and internal friction angle ( $\phi$ ) values.

# 3.11. Mineralogy

The minerals identified in the soils using X-ray diffraction are shown in Table 3. Soils that contain swelling clays minerals have been reported to have deleterious effects on the optimum utilization of these soils in engineering works.

Sample symbol	Major minerals identified	Minor minerals identified
MCS	Quartz, Kaolinite, Illite	Hematite
QTZ	Quartz, Kaolinite	Feldspar
MIG	Quartz, Kaolinite	Feldspar, Goethite
NPS	Quartz, Kaolinite,	Hematite
AMP	Quartz, Kaolinite,	Hematite, Goethite

Table 3. Summary of major and minor mineral identified in the samples



Figure 4. Typical x-ray diffractogram of the soil samples (MCS)

The dispersivity of a soil is directly related to its clay mineralogy <sup>[23]</sup>. The presence of dispersive soils in the embankment dam or foundation has been said to be one of the factors contributing factor to failures in structures which were reasonably well engineered <sup>[26-27]</sup>. Soils containing clay minerals such as montmorillonite tend to be dispersive, while those with kao-linite and related minerals (halloysite) are non-dispersive. Soils with illite tend to be moder-ately dispersive <sup>[24]</sup>. The xrd result shows that the analyzed five soil samples contain no undesirable mineral constituents except for MCS, which has illite as one of its major minerals, therefore making the soil less suitable in engineering works, particularly in dam construction due to its high swelling potential.

# 4. Conclusions

This study has investigated the principal engineering properties of lateritic soils derived from quartzite/quartz-schist, Mica Schist, Amphibolite, Nupe Sandstone and migmatite gneiss

in parts of south-western Nigeria for their usability as engineering construction materials. Based on the geotechnical test results, AMP, NPS, QTZ and MIG soils could be used in dam and embankment construction, liners in landfill system, foundation and could also support the construction of drainage. MCS showed tendency to swell, shrink and disperse due to the presence of illite. Also, it can be inferred that the soils are limited in their potential for application as base course material in road construction due to their low values of CBR. However, the soils are still useable as subgrade materials. The soil properties such as permeability and strength generally reduces in soil samples with high amount of clay minerals. Furthermore, the suitability of the soil sample derived from different rock types in civil construction works is greatly influenced by the presence and type of clay mineral present.

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To whom correspondence should be addressed: Professor Olusegun Omoniyi Ige, Department of Geology and Mineral Sciences, University of Ilorin, Private Mail Bag 1515, Ilorin, Kwara State, Nigeria, E-mail: vickyige2002@yahoo.com; olusegun@unilorin.edu.ng

# Article

# **Open Access**

Design of Novel Synthetic Iron Oxide Nano-Catalyst over Homemade Nano-Alumina for an Environmentally Friendly Fuel: Experiments and Modelling

Aysar T. Jarullah<sup>1\*</sup>, Ban A. Al-Tabbakh<sup>2</sup>, Mustafa A. Ahmed<sup>1</sup>, Shymaa A. Hameed<sup>1</sup>, Iqbal M. Mujtaba<sup>3</sup>

<sup>1</sup> Chemical Engineering Department, College of Engineering, Tikrit University, Iraq

<sup>2</sup> Petroleum Research & Development Center, The Iraqi Ministry of Oil / Baghdad, Iraq

<sup>3</sup> Chemical Engineering Department, Faculty of Engineering & Informatics, University of Bradford, Bradford BD7 1DP, UK

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#### Abstract

Achieving an environmentally friendly fuel with respect to minimum sulfur compounds has recently became a significant issue for petroleum refining industries. This paper focuses on investigating oxidative desulfurization (ODS) process for removal of sulfur compounds found in light gas oil (LGO) in a batch reactor (at different reaction temperatures and batch time) using a novel nano-catalyst based on 4% iron oxide (Fe<sub>2</sub>O<sub>3</sub>) as an active component. Precipitation and Impregnation methods are used to prepare the nano-gamma alumina ( $\gamma$ -alumina) and to generate the new synthetic homemade nanocatalyst. A mathematical model is formulated for the ODS process to estimate the optimal kinetic parameters within gPROMS package. An excellent consistency with the experimental data of all runs with error less than 5% have obtained. The optimization results display that the new nanocatalyst prepared here is effective in removing more than 97% of the sulfur compounds from LGO resulting in a cleaner fuel.

Keywords: Nano-catalyst; Parameters estimation; Iron oxide; Gamma alumina; Mathematical model.

#### 1. Introduction

In recent years, growing global energy demand, strict environmental legislations on transportation fuels and depleting oil reserves have together formed a triangle of constraints posing great challenges to refiners. Growing emissions in the form of SO<sub>x</sub> will increase with increasing energy demand due to combustion of fuels in transportation or in oil refineries. These emissions are harmful because of the emitted sulfur dioxide (SO<sub>x</sub>) can react with water in the atmosphere forming acidic rains that harmful to soil, buildings, forests and ecosystems <sup>[1-3]</sup>. Also, sulfur emissions lead to aggravate heart illness, respiratory illnesses, trigger asthma and contribute to the formation of atmospheric particulates <sup>[4]</sup>.

Sulfur compounds in fuel oil can be classified into four main groups mercaptans, thiophenes (TH), benzothiophenes (BT), and dibenzothiophenes (DBT) <sup>[5]</sup>. As a result, environmental regulations on the sulfur level are legislated in transportation fuels to reduce the sulfur content in diesel to less than 10 ppm since 2005 in Europe, less than 15 ppm since 2006 in the US, and less than 50 ppm since 2008 in Beijing and Shanghai in China and recently less than 10 ppm <sup>[6]</sup>. Therefore, desulfurization of fuels is very essential process in petroleum industry, and there is a need to find new methods that are more efficient, cost effective meeting the expectations of environmental regulations and refining requirements <sup>[7]</sup>. Many methods have been applied to remove sulfur compounds from fuel oil, such as hydrodesulfurization (HDS), extractive distillation, selective adsorption, biodesulfurization, and oxidative desulfurization (ODS) <sup>[8]</sup>. Hydrodesulfurization (HDS) is a common method for sulfur removal, which is used in petroleum refineries, but this process has drawbacks such as higher investment costs, high

operating conditions and low efficiency on DBT <sup>[9-12]</sup>. Therefore, researchers focus on the nonconventional low cost and low severity processes. So, ODS process is regarded as an alternate technique and considered to be a good choice among them. ODS is considered as a promising desulfurization technology because it can be operated at low temperature and low pressure and does not require the use of hydrogen.



Figure 1. Schematic representation of sulfur removal in the ODS process

Also, ODS process can easily remove the refractory sulfur compounds owing to their high electron density <sup>[13-16]</sup>. ODS process can enhance the efficiency of sulfur removal without destroying and poisoning catalyst <sup>[17]</sup>. As shown in Figure 1, the sulfur compounds in fuel oil are oxidized to their respective sulfones or sulfoxides by oxidizing agents. These compounds formed can be easily removed from fuel oil by extraction or adsorption due to their high polarity <sup>[11, 18]</sup>.

The preparation of the catalyst and the choice of oxidant are also considered the main effective factors on the ODS process <sup>[19]</sup>. In the former works, several types of oxidants have been studied, such as  $O_2$ ,  $H_2O_2$  and organic peroxides <sup>[20-22]</sup>. The catalyst plays a very

important key role in the ODS process owing to its responsibility for activating oxidants <sup>[23]</sup>. The catalytic system can be classified into supported and non-supported catalyst.

Supported catalyst consists of support and active metal such as Mn/Al<sub>2</sub>O<sub>3</sub> <sup>[24]</sup>, Co-Mo/Al<sub>2</sub>O<sub>3</sub> and Ni-Mo/Al<sub>2</sub>O<sub>3</sub> <sup>[25]</sup>. Nano-catalyst can be used in this process where nano-structured materials having much higher activities than that of the corresponding bulk materials <sup>[26]</sup>. Catalytic activity increases with decreasing the size of the particles <sup>[27]</sup>. By reducing the domain size of catalyst particle as far as possible, the number of active sites can be maximized <sup>[28]</sup>. Alumina is widely employed as a catalyst or catalyst support in several chemical processes, such as ammonia synthesis, synthesis gas and hydrogen production, oils hydrogenation, petroleum refining, automotive emissions control, and others <sup>[29-30]</sup>. Comparing micron-sized alumina particles, nano alumina have many advantages such as high hardness, good wear resistance and outstanding mechanical properties at high-temperature <sup>[31-33]</sup>. A smaller particle size provides a much larger surface area for molecular collisions and hence increases the rate of reaction, making it a better catalyst and reactant <sup>[33]</sup>. Iron oxide is used in the oxidative desulfurization process as an active metal in catalyst due to it is inexpensive as compared to other metal oxides, and it has excellent physicochemical properties, such as porosity, high surface and electropositivity <sup>[34]</sup>.

The novelty of this study is to prepare a new homemade nano-catalyst for deep oxidative desulfurization process, which contain  $Fe_2O_3$  as active metal and  $\gamma$ -alumina as support. The loading of  $Fe_2O_3$  on  $\gamma$ -alumina has not been reported in the public domain for ODS process. Also,  $\gamma$ -alumina nanoparticles used as a support of the catalyst is prepared by precipitation method from simple available raw material in addition to the precipitation method that has not been used to prepare  $\gamma$ -alumina as catalyst support for ODS process. As well as, oxidative desulfurization process is carried out by using real light gas oil (LGO) fraction as feedstock in a batch reactor utilizing the air as an oxidant.

The study aims to develop new homemade nano-catalyst for deep oxidative desulfurization process of light gas oil with low cost and less environmental effect via laboratory experiments. The objectives of this study can be summarized as follows:

- Preparation of homemade γ-alumina nanoparticles as support for catalyst.
- Preparation of new nano-catalyst, which is 4% Fe<sub>2</sub>O<sub>3</sub> /  $\gamma$ -alumina nanoparticles.

- Investigating the activity of the prepared catalyst via oxidative desulfurization reactions using light gas oil (LGO) fraction as feedstock at different operating conditions in a batch reactor utilizing the air as an oxidant.
- Studying the impact of the main operating variables (temperature and batch time) on the efficiency of desulfurization of the light gas oil.
- Determining the optimal kinetic parameters of the relevant reactions that can be used with high confidence to reactor design via optimization technique (two ways are employed for this purpose (linear and non-linear)).
- Achieving an environmentally friendly fuel by applying the optimization process to get the minimum sulfur content based on the validated model of the prepared nano-catalyst over homemade nano-  $\gamma$ -alumina.

## 2. Experimental work

## 2.1. Catalyst preparation

#### 2.1.1. Material

The chemical compounds that used for support and catalyst preparation are explained in Table 1.

Chemicals	Formula	Molecular weight, g/gmol	Purity %	Supplier
Ferric nitrate hydrate	$Fe(NO_3)_3.9H_2O$	404	98	Himedia
Aluminum nitrate	(Al(NO <sub>3</sub> ) <sub>3</sub> .9H <sub>2</sub> O)	375.13	97	GCC
Ammonium hydroxide	(NH4OH)	17.03	28%	GCC
Ethanol	C <sub>2</sub> H <sub>5</sub> OH	46.07	98	Hayaman

Table 1. List of materials and chemicals utilized for support and catalyst preparation

## 2.1.2. γ-Alumina preparation

In previous studies, the commercial  $\gamma$ -alumina has used as a catalyst support in ODS process <sup>[25-26]</sup>. In this study, the preparation process of  $\gamma$ -alumina is carried out by precipitation method that has the advantages of high purity, low cost, simple equipment manufacturing, large amount of production and short process beside available raw materials used in this method. So,  $\gamma$ -alumina is prepared here by the such method based on simple available raw material and simple equipment manufacturing.

A weight of (80 g) of Al(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O is dissolved in 100 mL of deionized water. Under constant magnetic stirring and in the presence of an ice bath, drops of ammonium hydroxide are added until the solution mixture is turned to foaming solution. The pH of the solution mixture is initially recorded at 2, then gradually increased and raised sharply from 2 to 7.5 during producing  $\gamma$ -alumina. Then, (50 mL of water + 50 mL of ethanol) is mixed and added to the foaming solution to remove any insoluble impurity. The filtered foaming solution is dried by using the furnace at temperature of 120°C for 12 hours. Such behavior causes the foaming solution to transfer to the solid state until the shrinkage in volume and changing in color of the foaming solution from grey to white is noticed. The dried sample is then crushed by hand in mortar to dispose the agglomeration of particles. After that, the samples is calcinated at a temperature of 650°C for 6 hours by using a furnace to obtain the nano  $\gamma$ -alumina and water as illustrated in equations 1 to 4 <sup>[35-36]</sup>:

$$Al(NO_3)_{3 (S)} + H_2O \longrightarrow Al(OH)_{3 (S)} + 3 NH_4NO_3 + H_2O$$
(1)

$$2Al(OH)_{3 (S)} \xrightarrow{110^{\circ}C} Al(OH)_{3 (S)} + 3 NH_4NO_3 + H_2O$$

$$(2)$$

$$2AIO.OH_{(S)} \xrightarrow{650^{\circ}C} \gamma - Al_2O_{3(S)} + H_2O$$
(3)

The process of calcination is carried out at steps where the temperature is increased with rate 6°C/min to 250°C for 1 h then to 450°C for 1 h and finally to 650°C for 4 h. The powder is cooled down until room temperature by switching off the furnace. The next step is crashing the powder, where the utilization active  $\gamma$ -alumina is crashed using a mortar and hammer to obtain final result of the nano activated  $\gamma$ -alumina. The prepared  $\gamma$ -alumina steps are shown in Figure.2.



Figure 2. γ-Alumina preparation steps

## 2.1.3. Preparation of Fe<sub>2</sub>O<sub>3</sub>/ γ-alumina catalyst

Homemade nano-catalyst (4% Fe<sub>2</sub>O<sub>3</sub>/ $\gamma$ -alumina) that has not been reported in the public domain in ODS process is prepared here by impregnation method. 0.95 gm of ferric nitrate hydrate is dissolved in 23 mL of deionized water then the solution is stirred by using magnetic stirrer for one hour at room temperature to get the saturated solution. 7.125 gm of the prepared  $\gamma$ -alumina nanoparticles are weighed in a beaker and the solution of ferric nitrate is added to the prepared  $\gamma$ -alumina in the beaker and stirring via magnetic stirrer for one hour at room temperature until the solution is completely impregnated. The impregnated  $\gamma$ -alumina is dried and calcinated in the furnace with four step processes. The solution is charged into beaker and placed in the furnace where temperature is raised to 120°C overnight. After that, the temperature is increased to 400°C for 2 h and finally to 550°C for 3 h to be gradually reduced. The purpose of calcination step is to convert metal salts loaded on  $\gamma$ -alumina to their corresponding metal oxides and deposition of the active metal oxide on the catalyst support. Also, the catalyst has acquired the physical and chemical properties. The prepared catalyst steps are shown in Figure.3.





## 2.2. Oxidative desulfurization reactions

## 2.2.1. Oil feedstock

Light gas oil (total sulfur content = 0.7510 wt %) obtained from KAR refinery/North of Iraq (Erbil) is utilized as liquid feedstock for ODS reaction. The physical properties of the light gas oil are illustrated in Table 2 and tested by the central laboratory of petroleum/ Erbil.

Table 2. Properties of light gas oil feedstock

Physical property	Values	Physical property	Values
Specific gravity at 15.5°C	0.8207	Pour point (°C)	<-20
API gravity	39.23	Distillation	(oC)
Total sulfur content (wt %)	0.7510	Initial boiling point (°C)	195
Kinematic viscosity at 313 K	3.21	10%	212
Flash point, (°C)	73	50%	246
Cetane index	54	90%	300
Cetane number	54	Final boiling point (°C)	328

## 2.2.2. Batch reactor

The oxidation reaction of sulfur compound is conducted in a batch reactor. Three necks round bottom flask of 500 mL is used for the reaction.



The middle neck is connected to a vertical condenser to condense the vapor of oil feedstock in order to permit air to leave only. The second neck used as air inlet which connected to compressor and the air will reach to the bottom of the flask by glass tube while the third neck is used to measure the temperature in the flask by inserting a thermometer to the solution inside the flask and to withdraw the sample reaction when the time is approached. The heating and mixing of the batch reactor is carried out via heating mantle stirrer. The process diagram and experimental device of ODS is shown in Figure 4.

Figure 4. Process diagram of batch reactor system

## 2.3. Experimental testing

## 2.3.1. Operating conditions

In this work, the experimental work includes several runs with respect to ODS process based on the following operating conditions:

- Reaction temperature: 90°C, 110°C, 130°C, 150°C.
- Batch time: 40min, 60min, 80min, 100min.
- The air flow rate is 120 L/h and the pressure is 1atm.

## 2.3.2. Oxidation of sulfur compounds

The oil feedstock is light gas oil containing sulfur compound. The prepared catalyst and the device are installed to run all the experiments and the following steps are performed in each run:

- 80 mL of feedstock is charged to the round bottom flask.
- The flask has been placed in the heating mantle stirrer and connected to the air tube and condenser. It is confirmed that the cooling water is flowing through the condenser to prevent any evaporation of the light gas oil. A thermometer is inserted to measure the reaction temperature.
- When the required temperature is achieved, 0.8 g of the prepared catalyst is added to the reactor with a compressor operation and the time is recorded.

- At the end of the run, the heating mantle stirrer is turned off and the product material is filtered.
- The reactor is washed, drying the process and prepare to start the next run.
- All products are tested for sulfur content.

#### 2.3.3. Adsorption of oxidized sulfur

The adsorption process between catalyst and sulfur compounds includes the contact of sulfur compounds that have low negative charge on the surface of catalyst having low positive charge and forming a polar interaction with the surface of catalyst where the oxidation reactions occur. So that, oxidation process increases the polarity of the oxidized sulfur compounds leading to increase the selectivity of sulfur compounds toward adsorption process due to such polarity [37].

#### 2.4. Analysis of liquid samples

The sulfur content in the feedstock and products has been tested in laboratory of oil and its derivatives- Erbil/Iraq. Product sulfur content is analyzed by X-ray fluorescence following the ASTM D4294 method.

#### 3. Mathematical model of batch reactor for ODS reaction

Mathematical modeling is the art of translating problems from an application area into tractable mathematical formulations whose theoretical and numerical analysis provides insight, answers, and guidance useful for the originating application <sup>[38]</sup>. The model used in this study is composed by a set of equations taken from literature while the kinetic parameters are estimated by reducing the sum of the squared error between experimental data and model prediction. The gPROMS (general Process Modeling System program) is used for modeling, simulation and optimization of ODS process based on the prepared catalyst <sup>[39]</sup>.

#### 3.1. Mass balance equations

Mass balance equation of batch reactor for ODS process is composed of several differential and algebraic equations. The general mass balance over catalytic batch reactor in total sulfur compound inside reactor is

Accumulation=Input-Output+Generation or disappearance by chemical reaction	(4)
Input= Output = 0 (in the batch reactor)	(5)
Accumulation of sulfur = $V.\frac{dC_{sulfur}}{dt}$	(6)
Disappearance of sulfur by the reaction = $V.(-r_{sulfur})$	(7)
Substitution of Eq. (5) into Eq. (4) giving the following eq.	
Accumulation = Disappearance by reaction	(8)
From Equations (6 to 8)	
$\frac{d_{sulfur}}{dt} = (-r_{sulfur})$	(9)
$dt = \frac{d_{sulfur}}{dt}$	(10)
(-r <sub>sulfur</sub> )	()
where: at $t=0 C_{sulfur} = C_{sulfur,t}$ ; at $t=t C_{sulfur} = C_{sulfur}$ .	
By integration:	
$t = \int_{c}^{C_{sulfur}} \frac{d_{sulfur}}{d_{sulfur}}$	(11)
<sup>2</sup> C <sub>sulfur,t</sub> (-r <sub>sulfur</sub> )	、 /

#### 3.2. Chemical reaction rate

The mechanism and the kinetic of catalytic air oxidation of sulfur are complex involving many steps to reach the end of reaction. The complexity of the chemical reaction could be taken into account by assuming nth order kinetics.

$$(-r_{sulfur}) = K_{ann}C_{sulfur}$$

(12)

The apparent kinetic constant  $(k_{app})$  is associated to intrinsic kinetic constant by internal diffusion that is represented by the catalyst effectiveness factor  $(\eta_0)$  <sup>[40]</sup>. So that, the reaction rate equation can be presented as follow:

$$\left(-r_{sulfur}\right) = \eta_0 \, k \, C_{sulfur}^{n}$$

(13)

The reaction rate constant is influenced by the temperature according to (Arrhenius equation) [41-42].

$$k = k_0 e^{\left(-\frac{EA}{RT}\right)}$$
(14)  
From equation (13 and 14),  
$$\left(-r_{sulfur}\right) = \eta_0 k_0 e^{\left(-\frac{EA}{RT}\right)} C_{sulfur}^{n}$$
(15)

Also, ko and EA can be calculated by linearization of equation 14, which gives the following equation:  $lnk = lnk_o - (\frac{-EA}{R}\frac{1}{T})$ (16)

By substitute equation 13 in equation 11 and integration these equation, the final expression of the catalytic oxidation reaction of sulfur with n<sup>th</sup> order kinetic can be presented as follows:

$$C_{sulfur} = [C_{sulfur,t}]^{(1-n)} + (n-1).t.K_{app}]^{(1-n)}$$
(17)

#### 3.3. Reactor performance

The oxidation reaction of the sulfur compound present in light gas oil is carried out in the batch reactor. The process includes number of parameters mainly, diffusivities, effectiveness factors, oil viscosity and others. These parameters are determined by using the correlations presented in this chapter.

 $K_{app} = \eta_0 K_{in}$ 

Hence, equation 17 is written as follows:

$$C_{sulfur} = \left[C_{sulfur,t}^{(1-n)} + (n-1).t.K_{in}.\eta_0\right]^{\left(\frac{1}{1-n}\right)}$$
(19)

## 3.3.1. Effectiveness factor (ηο)

The effectiveness factor  $(\eta_0)$  can be determined as function of Thiele modulus with the following equation valid for sphere particles [43-44].

 $\eta_0 = \frac{3(\phi \coth \phi - 1)}{12}$ φ<sup>2</sup>

While, the generalized Thiele modulus for nth-order irreversible reaction is determined by the following equation [43-44].

$$\Phi = \frac{V_P}{S_P} \sqrt{\left(\frac{n+1}{2}\right) \frac{k_{in} C_{sulfur}^{(1-n)} \rho_P}{D_{ei}}}$$
(21)

## 3.3.2. The effective diffusivity (D<sub>ei</sub>)

٦	he effective	diffusivity of	the catalyst st	ructure (porosit	y and tortu	osity) is re	presented by	,
taki	ng the pores	network insi	de the particle	into account a	s follows <sup>[40</sup>	,44].		

 $\frac{1}{D_{ki}}$ where, catalyst porosity ( $\mathcal{E}_{B}$ ) can be calculated by the following two equations based on experimental data:

$$\begin{aligned} & \mathcal{E}_B = V_g \rho_p \\ \rho_p = \frac{\rho_B}{1 - \varepsilon_P} \end{aligned} \tag{23} \end{aligned} \tag{24}$$

The tortuosity factor ( $\mathcal{T}$ ) of the pore network have the value of (2 to 7)<sup>[45]</sup>. According to literatures, the tortuosity factor has reported to be 4 [40-43]. The effective diffusivity within the catalyst particle contains two types of diffusivity, Knudsen diffusivity Dki and molecular diffusivity D<sub>mi</sub>.

The Knudsen diffusivity is calculated as follows [40,43]:	
$D_{ki} = 9700 r_g (\frac{T}{M_w})^{0.5}$	(25)
where, mean pore radius $(r_q)$ is estimated from this equation [46]:	
$r_g = \frac{2V_g}{S_g}$	(26)
The molecular diffusivity is calculated by Tyn-Calus equation [47-48]:	
$D_{mi} = 8.93 * 10^{-8} \left( \frac{v_l^{0.267} T}{v_s^{0.433} \mu_l} \right)$	(27)

(18)

(20)

(22)

## 3.3.3. Molar volume

The molar volume of the model sulfur compound is calculated by the following equation <sup>[48]</sup>:

$v_{sulfur} = 0.285 (v_{csulfur})^{1.048}$	(28)
$v_l = 0.285 (v_{cl})^{1.048}$	(29)
$v_{cl} = (7.5214 * 10^{-3} (T_{meABP})^{0.2896} (\rho_{L15.6})^{-0.7666}) M w_L$	(30)

The critical volume of liquid (light gas oil) is estimated by a Riazi-Daubert correlation <sup>[49]</sup>:

#### 3.3.4. External volume( $V_p$ ) and surface ( $S_p$ ) of the catalyst

The external volume (V\_p) and external surface (S\_p) of the catalyst can be calculated according to shape of the particle; for sphere particle:

$V_p = \frac{4}{3}\pi (r_p)^3$	(31)
$S_n = 4\pi (r_n)^2$	(32)

## **3.3.5.** *Viscosity*(µ<sub>l</sub>)

The viscosity of light gas oil can be calculated by using Glaso's equation as follows <sup>[50]</sup>:  $\mu_l = 3.141 * 10^{10}(T - 460)^{-3.444}(logAPI)^{\alpha}$  (33) Where, (a) is Dimensionless number and estimated from this equation:  $\alpha = 10.313[log_{10}(T - 460)] - 36.447$  (34) Also, American Petroleum Institute (API) is estimated from this equation:  $API = \frac{141.5}{sp.gr_{15.6}} - 131.5$  (35)

The equations of the mathematical model in this chapter are encoded for the ODS interaction and solved using the gPROMS (general Process Modeling System) package.

#### 4. Kinetic parameter estimation technique

Estimating the kinetic parameter is very important step in many fields of science and engineering when many physiochemical processes are described by the equations system containing unknown parameters. Recently, the benefits of developing kinetic models for chemical engineers have increased with accurate calculations of parameters due to advanced control techniques and optimization of process, which can apply the fundamental models.

The appropriate value of the kinetic parameters can be estimated by reducing the errors between experimental data and predicted data by the mathematical model. Therefore, the predicted values from the model should match the experimental data as closely as possible <sup>[51]</sup>. For the purpose of optimizing the process, reactor design, process control and catalyst selection, it is important to develop kinetic models that can accurately predict the concentration of the product under the conditions of the process.

In this study, the best values of the kinetic parameters of the relevant reactions are achieved by utilizing two approaches that depend on the sulfur content in the oxidation process under different operating conditions. These approaches are given below:

- Linear regression: Determines the reaction rate constant (k) and order of the interaction (n), then utilizing the Arrhenius equation with linear regression to evaluate the activation energy (EA) and pre-exponential factor (k₀).
- Non-linear regression: Evaluates the order of reaction (n), the pre-exponential factor  $(k_o)$  and activation energy (EA) directly.

In order to estimate the best value for the kinetic parameter, the following objective function was minimized as shown below:

$$0BJ = \sum_{n=1}^{Nt} \left( C_{sulfur}^{exp} - C_{sulfur}^{pred} \right)^2$$

(36)

In equation (36), Nt,  $C_{sulfur}^{exp}$  and  $C_{sulfur}^{prd}$  represent the numbers of runs, the experimental concentration and predicted concentration by model of sulfur content respectively. The conversion of sulfur compound can be calculated using the following equation:

$$X_{sulfur} = 1 - \frac{c_{sulfur}}{c_{sulfur,t}}$$

(37)

#### 4.1. Optimization problem formulation for parameter estimation

The formulation of optimization problem for parameter estimation can be stated as follows: **Given:** The reactor configuration, the catalyst and the process conditions.

**Obtain:** For the first approach: The reaction orders of oxidation reaction (n) for the catalyst and reaction rate constant (k) at different temperature (363, 383, 403, 423) K respectively and then calculation the activation energy and pre-exponential factor by linear regression by Arrhenius equation. For the second approach: the reaction order (n), activation energy (EA) and pre-exponential factor (ko) are simultaneously estimated for the catalyst.

**So as to minimize:** The sum of squared error (SSE).

**Subjected to:** Constraints on the conversion and linear bounds on all optimization variables Mathematically using linear regression, the optimization problem can be presented as follow: **Min: SSE** 

 $\begin{array}{l} n, k_i, (i = 1 - 4) \\ \text{S.t.f}(z, x(z), \dot{x}(z), u(z), v) = 0 \\ C_L \leq C \leq C_U \end{array}$ 

 $n_l \le n \le n_u$ 

 $k_{iL} \leq k_i \leq k_{iU}$ 

While by using the second approached (nonlinear regression) the problem can be presented as follow:

Min:

#### SSE

 $n, EA, k_0$ S.t.f(z, x(z),  $\dot{x}(z)$ , u(z), v) = 0  $C_L \le C \le C_U$  $n_l \le n \le n_u$  $EA_L \le EA \le EA_U$  $k_{OL} \le k_i \le k_{OU}$ 

S.t.f(z,  $x(z),\dot{x}(z), u(z),v$ ) = 0: represent the process model that presented previously.

z: is independent variable; u(z): is the decision variable; x(z): represent the set of all variables;  $\dot{x}(z)$ : represent the derivative of the variables with respect to time; v: is the design variable; C, C<sub>L</sub>, C<sub>U</sub>: concentration, lower and upper bounds of concentration; L, U: are lower and upper bounds.

The method of optimization solution by gPROMS is performed by two steps that can be presented as follows <sup>[52]</sup>:

- First, performs a simulation which converge all the equality constraints described by function (f) and also to satisfy the constraints of inequality.
- Secondly; performs the optimization (the values of the decision variables such as the kinetic parameters that can be updated).

## 5. Results and discussion

## 5.1. Catalyst characterization

#### 5.1.1. Chemical composition

XRF is used to determine the structural formula of the prepared samples and the actual metal oxide loaded (Fe<sub>2</sub>O<sub>3</sub>). As shown in Table 3, the purity of the prepared  $\gamma$ -alumina is 97.8%. The chemical composition for the prepared catalyst are summarized in Table 4. A good percentage of the active metal appear is observed.

Compounds	Concentration
Al <sub>2</sub> O <sub>3</sub>	69.07
SiO <sub>2</sub>	0.804
P <sub>2</sub> O <sub>5</sub>	0.508
SO <sub>3</sub>	0.021
Other components	0.227
Sum of concentration	70.63

Table 4. Chemical composition of the prepared catalyst

Compounds	Concentration
γ-Al <sub>2</sub> O <sub>3</sub>	94.14
Fe <sub>2</sub> O <sub>3</sub>	4.117

## 5.1.2. X-ray diffraction (XRD)

Figure 5 shows the XRD pattern of synthesized nano  $\gamma$ -alumina powder prepared. The three main reflections of nano  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> phase are obviously observed as broad peaks at 20 angles around (36.8263°, 45.8903°, and 66.6322°), respectively <sup>[53-55]</sup>. The peaks in the pattern are significantly indicated that the formation of nano sized  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> crystallites is obtained. The size of crystallites was correlated using Scherrer's equation <sup>[56]</sup> as follows.

 $d = 0.94\lambda/\beta \cos \Theta$ 

(38)

where d is the diameter of crystallite;  $\lambda$  is the X-ray wave length;  $\beta$  is the broadening line at the half maximum intensity which represents the full width at half maximum (FWHM);  $\Theta$  is the Bragg angle at which the scattering wave was reflected or scattered at lattice plane producing intense peaks.



Figure 5. XRD patterns for synthesized nano  $\gamma$ -alumina powder

The average diameter of crystallite is 27.32 nm for the prepared nano  $\gamma$ -alumina powder. The percent crystallinity of samples is ascertained by comparing the ratio of intensity of the peaks for the prepared  $\gamma$ -alumina with the corresponding ratios of standard  $\gamma$ -alumina sample <sup>[57]</sup>.

Table 5. characteristic peaks and their relative intensities for prepared nano  $\gamma\text{-alumina}$  powder

20	intensity I	intensity I standard	
66.6322	100	100	
45.8903	83	80	
37.7555	27	65	

Therefore, crystallinity % is determined as follows [52]:



(39)

The percent crystallinity is estimated to be 85.7% for prepared nano  $\gamma$ -alumina powder. The crystallinity is considered to be one of the most important effected factors on thermal stability, where, thermal stability is increased with increasing the crystallinity <sup>[58-59]</sup>. Figure 6 shows the XRD patterns of the prepared catalyst after calcination at 550°C. The

Figure 6. XRD patterns for the prepared catalyst diffractogram of commercial iron oxide shows the characteristic peaks of  $Fe_2O_3$  at 33.2°, 35.6°, 49.4°, 54.1°, 62.4° and 63.9° <sup>[60]</sup>.

When impregnated with  $Fe_2O_3$  in the catalyst, it is seen that the intensity of the peaks corresponding to  $\gamma$ -alumina drops, and the peaks corresponding to  $Fe_2O_3$  start appearing as shown in the Figure 6.

## 5.1.3. Surface area and pore volume analysis

The surface area, pore volume and pore size of synthesized support and catalyst are summarized in Table 6. It can be observed that after impregnation, pore volume and the surface area decreased slightly due to the occupation of the active component in some spaces within the samples. Also, The bulk density is 0.482 gm/cm<sup>3</sup> for the prepared catalyst.

Table 6. Summary of surface area, pore volume and pore size of the prepared  $\gamma$ -alumina and catalyst

Compounds	γ-alumina	Fe <sub>2</sub> O <sub>3</sub> /γ-alumina	
Surface area (BET), m <sup>2</sup> /g	263.91	256.68	
Pore volume, cm3/g	0.3163	0.316	
Pore size, nm	4.79419	4.92913	

#### 5.1.4. Fourier transform infrared (FTIR)

FTIR analysis for the synthesized  $\gamma$ -alumina in the wave number region of 4000-500 cm-1 are shown in Figure 7. The band located at 3412 cm<sup>-1</sup> was attributed to the O–H stretching vibration and the band at ~1650 cm<sup>-1</sup> was related to the H–O–H symmetric stretching vibration of adsorbed water molecules. The absorption bands centered at 650 attributed to stretching vibrations of Al-O, which is characteristic of  $\gamma$ -alumina as shown in Figure 7 <sup>[61-63]</sup>. Figure 8 shows the FTIR spectra of the synthesized nano catalyst after loading of Fe<sub>2</sub>O<sub>3</sub>. Based on the results presented in Figure 7 and 8, there is no difference in the wave number regions before and after loading. Such good results indicated that there is no chemical change in the composition of the support after loading process. Also, no bonds of iron oxides were found resulting that there is insufficient amount of iron oxide to give the peaks.



Figure 7. FTIR for synthesized  $\gamma$ -alumina

5.1.5. Thermal gravimetric analysis (TGA)



Figure 9. TGA of synthesized (a) nano  $\gamma$ -alumina, (b) catalyst



Figure 8. FTIR for synthesized catalyst

Figure 9 shows the TGA profiles of the prepared  $\gamma$ -alumina and catalyst samples. From this Figure, there are two regions of mass loss where, the first one, (between 20-120°C), with mass loss values around 10 %, is related to the evaporation of physically adsorbed water on the solid <sup>[64-65]</sup>. In the range of 180-350°C, it comes from the release of the chemisorbed water, which takes place around 250°C <sup>[66]</sup>. At temperatures above 450°C, the weight of all samples tends to remain stable. The mass loss values observed

in the TGA experiments is -20.84% for the prepared  $\gamma$ -alumina and -26.10% for the catalyst. Such good results indicated that the support and prepared catalyst have a good thermal stability.

## 5.1.6. Particle size distribution

Figures 10 shows the particle distribution of the prepared catalyst. The average nano particle size is 84.65 nm and the overall range of the diameters between 55 - 135 nm. Maximum volume percentage of the particles is 23.76 % at size distribution of 90 nm, and minimum volume percentage is 1.03 % for particles at size of 130 nm.



Figure 10. Granularity accumulation distribution of the synthesized catalyst

# 5.1.7. Scanning Electron Microscopy (SEM)

As shown in Figure 11, the SEM of the catalyst showed that the particle size is <100 nm. Also, the shape of the particles that are presented in these Figure are in the spherical form.



Figure 11. SEM images of the synthesized catalyst

# 5.2. Oxidative desulfurization results

The oxidative desulfurization process is employed to test the activity of the prepared catalyst. Light gas oil (LGO) is used as an oil feedstock and air as an oxidant in a batch reactor. Several operating conditions affecting the reduction of sulfur content such as reaction time and reaction temperature are studied.

## 5.2.1. Effect of reaction temperature

The impact of the reaction temperature on the sulfur removal in ODS reactions has studied at 363 K, 383 K,403 K and 423 K, and the results are illustrate in Figure 12.


Figure 12. Temperature influence of sulfur removal for the prepared catalyst at various reaction time

It can be observed from Figure 12 above that increasing the reaction temperature leads to increase the conversion of sulfur compound. Such attitude is due to the fact that the temperature affect positively the reaction rate constant (rate constant) leading to an increase in the sulfur compound conversion <sup>[67]</sup> according to Arrhenius equation. So, increasing the temperature causes an increase in the number of molecules having activation energy and thus an increase in the number of molecules involved in the oxidation reaction resulting in an increase in conversion.

Also, the catalyst activities is highly affected by the calcination temperatures. The density of acid sites is strongly influenced by the catalyst calcination temperature; it is increased to reach a maximum that leads to enhance the removal of sulfur compound from diesel fuel. The optimum temperature for y-Al<sub>2</sub>O<sub>3</sub> powder synthesized by flash calcination is 650°C that surface area and pore volume are maximized <sup>[68]</sup>. So, this study was used the calcination temperature of  $650^{\circ}$ C to prepare y-Al<sub>2</sub>O<sub>3</sub> for achieving high catalytic activity in ODS process.

#### 5.2.2. Effect of reaction time



Figure 13. Time influence of sulfur removal for the prepared catalyst at various reaction temperature

The impact of the reaction time on the removal of sulfur compound by oxidation reaction has investigated at 40 min, 60 min, 80 min and 100 min, which are illustrated in Figure 13. In general, the results showed that an increase in the desulfurization efficiency with the reaction time. Where, increasing the time of the reaction offers the chance for the reactants to contact then to react among them. As a result, the contact time among the reactants increases giving longer contact with the active site of the catalyst [69]. In Figure 13, the conversion of sulfur compound increased from 50.33% to 70.29% when the reaction time increases from 40 min to 100 min at 423 K.

5.3. Kinetic parameters estimation

The optimal kinetic parameters can be estimated using a mathematical model aimed at minimizing the error between the experimental data and the predicted data by the mathematical model to get predicted values from model as close as possible to the experimental data <sup>[70]</sup>. The constant parameters used in the mathematical model are shown in Table 7.

Parameter	Symbol	Unit	Value
Initial concentration	Ct	wt%	0.751
Time	Time1,	min	time1=40, time2=60,
	time2,		time3=80, time4=100
	time3, time4		
Temperature	T1, T2, T3,	К	T1=363, T2=383, T3=403,
	T4		T4=423
Density of light gas oil at 15.5°C	ρι	gm/cm <sup>3</sup>	0.8205
Mean average boiling point	T <sub>meABP</sub>	°R	957

Table 7. Values of constant parameters used in ODS model

Parameter	Symbol	Unit	Value
Acceleration gravity	g	m/sec <sup>2</sup>	9.81
Gas constant	R	J/mole.ºK	8.314
Pore volume per unit mass of cat-	Vg	cm³/gm	0.316
alyst			
Specific surface area of particle	Sg	cm²/gm	2566800
Volume of catalyst particle	Vp	cm <sup>3</sup>	3.1759*10 <sup>-16</sup>
External surface area of particle	Sp	cm <sup>2</sup>	2.25114*10 <sup>-10</sup>
Bulk density	ρ <sub>в</sub>	gm/cm <sup>3</sup>	0.482
Molecular weight of light gas oil	M <sub>wL</sub>	gm/mole	200.468
Molecular weight of sulfur	Mwi	gm/mole	32.06
Mean pore radius	r <sub>g</sub>	nm	2.4645

#### 5.3.1. Linear regression

The optimal results of the model parameters obtained by linear approach are reported in Table 8 below for the catalyst:

Table 8: Optimal model parameters obtained by optimization process using linear approach

_		
Parameter	Value	Unit
n	1 0122122	
11	1.9133123	_
k1	0.00674264	$(wt^{-0.9133123}), min^{-1}$
	0.000.120.	(,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,
k2	0.01183655	$(wt^{-0.9133123})$ . $min^{-1}$
43	0 01921438	(wt = 0.9133123) min = 1
KJ	0.01721450	(we ).mun
k4	0.02902299	$(wt^{-0.9133123})$ . $min^{-1}$
SSE	1 802865×10-3	
55L	1.002003×10	_

### **Activation energy**



According to Arrhenius equation, a plot of (lnk) versus (1/T) gives a straight line with slope equal to (-EA/R) in which the activation energy is calculated as illustrated in Figure 14.

The activation energy and preexponential factor which can be obtained from this Figure is 31.093 kJ/mol and 203.365, respectively.

Figure 14. (In k) versus (1/T) for kinetic parameters of the oxidation process

#### 5.3.2. Non-linear regression

In this approach, pre-exponential factor, the activation energy, and interaction order, have simultaneously been estimated and the values of these parameters are shown in Table 9.

Table 9. Optimal kinetic model parameters obtained via optimization process by non-linear approach

Parameter	Value	Unit
n	2	_
EA	34.305	KJ/mol.
ko	578.64	$wt^{-1}$ . $min^{-1}$
SSE	1.712543×10 <sup>-3</sup>	_

# 5.4. Experimental and simulation results

The gPROMS software is used for the simulation of the process. The experimental and predicted results are summarized in Tables 10 and 11 below, and also illustrated in Figure 15 (for non-leaner approach).

Tempera-	Time	Sulfur content (wt %)		Convers	sion	
ture (K)	(min)	Experimental	Predicted	Experimental	Predicted	Error %
363	40	0.610	0.621	0.188	0.173	1.85
363	60	0.571	0.571	0.240	0.240	0.06
363	80	0.531	0.528	0.293	0.297	0.49
363	100	0.499	0.491	0.336	0.346	1.50
383	40	0.537	0.548	0.285	0.270	2.10
383	60	0.476	0.482	0.366	0.358	1.23
383	80	0.436	0.429	0.419	0.428	1.53
383	100	0.397	0.387	0.472	0.485	2.49
403	40	0.450	0.468	0.400	0.377	3.91
403	60	0.407	0.392	0.458	0.478	3.63
403	80	0.349	0.337	0.536	0.552	3.46
403	100	0.309	0.295	0.589	0.608	4.69
423	40	0.373	0.391	0.503	0.480	4.74
423	60	0.302	0.133	0.598	0.823	3.58
423	80	0.259	0.260	0.655	0.654	0.54
423	100	0.223	0.222	0.703	0.704	0.40

Table 10. Experimental and simulation results using linear approach

Tabla 11	Evennimental	and	cimulation	roculto	uning	non linoar	annraach
Table II.	Experimental	anu	SIIIIUIALIOII	results	usina	non-inear	approach

Tomporatura	Time	Sulfur conten	it (wt %)	Conversi	on	
(K)	(min)	Experimental	Predicted	Experimental	Pre- dicted	Error %
363	40	0.610	0.625	0.188	0.168	2.55
363	60	0.571	0.577	0.240	0.232	1.11
363	80	0.531	0.536	0.293	0.287	0.92
363	100	0.499	0.500	0.336	0.335	0.25
383	40	0.537	0.551	0.285	0.267	2.52
383	60	0.476	0.486	0.366	0.353	2.02
383	80	0.436	0.435	0.419	0.421	0.34
383	100	0.397	0.393	0.472	0.477	0.90
403	40	0.450	0.463	0.400	0.383	2.85
403	60	0.407	0.389	0.458	0.483	4.46
403	80	0.349	0.335	0.536	0.554	3.98
403	100	0.309	0.294	0.589	0.609	4.83
423	40	0.373	0.374	0.503	0.502	0.24
423	60	0.302	0.299	0.598	0.602	1.07
423	80	0.259	0.249	0.655	0.669	3.82
423	100	0.223	0.213	0.703	0.716	4.41

The sums of squared errors (SSE) is 0.0018 and 0.0017of linear approach and nonlinear approach respectively. So, the parameter estimated by the nonlinear approach is more accurate than those calculated by the linear approach. The values of the activation energy (EA) and pre-exponential factor ( $k_0$ ) estimated via linearization (linear approach) of Arrhenius equation gives high error as compared with those estimated via non-linear method (nonlinear approach).



Figure 15. Comparison between experimental and simulated data at (a) 40 min (b) 60 min (c) 80 min (d) 100 min

From these results, it is observed that increasing the reaction temperature and reaction time increases the reaction rate of ODS process based on the following points.

- **Temperature:** Increasing the reaction temperature leads to increase the conversion of sulfur compound due to the reaction rate constant that is influenced by the temperature. Where, the temperature is dependent by Arrhenius equation leading to an increase in the sulfur compound conversion. Also, rising the reaction temperature contributes in an increasing of magnitudes of some important physical properties such as diffusivity, viscosity and surface tension. Thus, the rate of absorption of molecular air into light gas oil and the diffusing rate of the sulfur compound beside the rate of dissolving air inside the pores of the catalyst to reach the active sites increases with increasing the reaction temperature.
- **Time:** The rise in the reaction time will increase the contact time among the reactants on the active sites of catalyst. So, high residence time is achieved giving high reaction.

### 6. Optimal operation conditions for maximum conversion of ODS process

#### 6.1. Optimization problem formulation for maximum conversion

Based on the experiments and after obtaining the optimal kinetic parameter for the ODS process, the optimal operating conditions for obtaining the minimum sulfur content are necessary. Therefore, the optimization problem can be formulated for the maximum conversion of the process as follows:

**Given:** The reactor configuration, the reaction order, the catalyst and pre-exponential factor and activation energy for the reaction.

**Obtain:** The best operating conditions for high conversion.

**So as to minimize:** The sulfur concentration.

**Subjected to:** Process constraints and linear bounds on all optimization variables in the process.

Mathematically, the problem can be represented as follows: **Min:** 

Min:  $C_{sulfur}$   $T, time_i, C_{sulfur}$   $S.t. f(z, x(z), \dot{x}(z), u(z), v) = 0$   $time_L \leq time \leq time_U$   $C_{sulfur,tL} \leq C_{sulfur,t} \leq C_{sulfur,tU}$   $T_L \leq T \leq T_U$  $X_{sulfur,tL} \leq X_{sulfur,t} \leq X_{sulfur,tU}$ 

The optimization solution method is performed by gPROMS software.

### 6.2. Optimal operating conditions for maximum conversion

After obtaining the optimal kinetic parameters, such optimal values will be used in the kinetic model in order to obtain the best operating conditions to give the minimum sulfur content in the products based on the prepared catalyst achieving the main goal of this study, which is an environmentally friendly fuel. The optimal values of the operating conditions for the prepared catalyst are presented in Table 12.

Table 12. Optimal operating conditions for ODS process

Parameter	Value	Unit
C <sub>sulfur.t</sub>	0.757	wt%
Т	550	К
Time	200	min
Conversion	97.97	%

The sulfur content in the petroleum products has been reduced by finding the optimal operating conditions to achieve the environmental and industrial aspects demands .As can be seen from the presented results in Table 12, the maximum sulfur removal (higher than 97%) has been achieved at the reaction temperature of 550 K, contact time at 200 min and initial concentration of sulfur compound at 0.757 ppm to match the environmental regulations to get almost free sulfur content and as a result high fuel oil quality.

### 7. Conclusions

In this study, the homemade nano catalyst ( $Fe_2O_3/\gamma-Al_2O_3$  nanoparticles) is successfully applied for ODS process using air as oxidant in batch reactor. The nano-gamma alumina is prepared by precipitation method and then the impregnation method is used to generate the catalyst. The method of impregnation is a good method in preparing the nano catalyst because it gives a good distribution of the active metals in addition to the high surface area and the distribution of pore for prepared catalyst. Several characterization tests such as SEM, XRD, XRF, TGA, FTIR, BET and particle size distribution were conducted on the prepared  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> and the prepared catalyst. Such tests indicated that  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> produced by precipitation method has high purity, high surface area and high crystallinity. Also, a good distribution of active metal (Fe), various surface morphology and high dispersion of active metal were obtained. As well as, the result of the particle size distribution test for the prepared nano-catalyst indicated that the particle size of the catalyst is reported to be < 100 nm. The experimental conversion of sulfur compounds presented in real light gas oil has (70.29%) under the conditions of temperature = 423 K and reaction time = 100 min. The simulation and optimization techniques are applied in this study to estimate the optimal kinetic parameter based on experimental results. Estimation of these parameters is conducted by minimizing the sum of squared error between experimental and predicted results and all results gave absolute error less than 5% at different conditions. The simulation and optimization techniques are achieved using two approaches (linear and non-linear method) and the simulation results show that the non-linear approach is more accurate due to it give less (SSE) in comparison with linear approach. The optimal kinetic parameters can be used to estimate the optimal operating conditions to achieve cleaner fuel with sulfur conversion above 97% at process temperature= 550 °K, batch time=200 min and initial concentration = 0.7574 wt%."

#### Nomenclature

$C_{sulfur}$	Sulfur Concentration
X <sub>sulfur</sub>	Conversion of sulfur compound.
C <sub>sulfur</sub>	Concentration of sulfur compound at the end of the reaction.
C <sub>sulfur.t</sub>	Initial concentration of sulfur present in light gas oil.
k	Reaction Rate Constant
$k_{App}$	Apparent Rate Constant
$\mathcal{D}_{ei}$	Effective diffusivity
$\mathcal{D}_{ki}$	Knudsen diffusivity
$\mathcal{D}_{mi}$	Molecular diffusivity
Sp. gr <sub>15.6</sub>	Specific gravity of of light gas oil at 15.6°C
EA	Activation Energy
$Mw_L$	Liquid molecular weight of
Mw	Molecular weight of sulfur
R	Gas constant
п	Order of reaction
$-r_{sulfur}$	Reaction rate of sulfur
$r_{g}$	Pore radius (nm)
$r_p$	Particle radius
$S_p$	External surface area of catalyst particle
$S_a$	Specific surface area of particle
V.	External Volume of catalyst particle
V	Pore volume
$f_g$	Mean average boiling point
<sup>1</sup> meABP	mean average boiling point

#### **Greek symbols**

- $\eta_0$  Effectiveness factor
- $\Phi$  Thiel modulus
- $\mathcal{E}_B$  Porosity
- T Tortuosity
- $\rho_B$  Bulk density
- $\rho_p$  Particle density
- $\rho_{L15.6} \qquad \text{Density of light gas oil at } 15.6^{\circ}\text{C}(\text{gm/cm}^3).$
- $\mu_l$  Viscosity of liquid
- $v_l$  Liquid molar volume
- $v_{cl}$  Critical molar volume of liquid
- $v_{Sulfur}$  Molar volume of sulfur
- *v*<sub>csulfur</sub> Critical volume of sulfur compound.
- <sup>o</sup> Initial (at time = 0)

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To whom correspondence should be addressed: Aysar T. Jarullah, Chemical Engineering Department, College of Engineering, Tikrit University, Iraq; <u>e-mail</u>: <u>A.T.Jarullah@tu.edu.iq</u>

# Article

Analysis, Modeling and Optimization of Reactors for Hydrotreatment of Diesel Fuel with Removing Organosulfur Impurities

#### Naum A. Samoilov

Ufa State Petroleum Technological University, Kosmonavtov St., 1, Ufa, Russia

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#### Abstract

The disadvantages of the reaction units of hydrotreatment plants are considered and an approach to process improvement by means of the characteristics of hydrodesulfurization reactions of organosulfur impurities of various nature is substantiated. The mathematical modeling method demonstrates that when dividing the feedstock into three wide fractions, each containing several narrow fractions, differential hydrotreatment of these wide fractions is ensured by a reduction in the catalyst load in the reactor unit of the facility.

Keywords: Hydrotreatment; Reactor; Diesel fuel; Pseudo-component; Mathematical modeling; Optimization.

#### 1. Introduction

Hydrotreating of diesel fuel refers to the most large-tonnage oil refining processes. The peculiarity of this process is its implementation in a three-phase system: liquid feed stock, hydrogen-containing gas and bifunctional solid catalysts which ensure hemolytic and heterolytic hydrogenation reactions of numerous organosulfur compounds that significantly complicates the formation of mathematical models of the process <sup>[1-5]</sup>. The process schemes of the interconnection of equipment at the hydrotreatment plants implemented in the industry are very conservative from the standpoint of the hardware design of the reactor unit and usually represent a complex of up to four reactors operating sequentially. The volume of hydrotreating catalysts in the reactor unit is determined by the amount of diesel fuel treated and its purification depth. The required catalyst loading is proportional to the capacity and increases dramatically (several times) with an increase in the purification depth. The design capacity of hydrotreating plants at the level of two million tons per year is achieved only with shallowtreatment with a sulfur content of 350 ppm in purified fuel used for roads and agricultural machinery. For vehicles (mainly cars) according to the Euro-5 standard, the sulfur content is limited to 10 ppm which is achieved by increasing the time of hydrotreatment and, as a result, reducing the capacity of hydrotreatment plants by 3 to 4 times or by construction of additional reactors during the revamping of plants.

The degree of activity of sulfur compounds in hydrogenolysis reactions is different and decreases in the series: mercaptans > sulfides > thiophenes > benzothiophenes > dibenzothiophenes. At the same time, the most difficult-to-hydrogenate compounds in the thiophene series are concentrated mainly in heavy fractions boiling above 330°C <sup>[6-9]</sup>. While ensuring the required quality of diesel fuel hydrotreatment, the purification process as a whole is precisely limited by the interaction with hydrogen of a relatively small amount of difficult-to-hydrogenate organosulfur, while the easily hydrogenated components have already undergone hydrogenolysis. In this situation, the assessment of the composition of the initial purified diesel fuel, which is necessary for the mathematical modeling of the process <sup>[6-7,10-11]</sup>, is of particular importance.

The identification of an extremely complete set of organosulfur components in diesel fuel and the development of a database of possible reaction pathways is practically impossible due to the scale of the task, high cost of the necessary reagents, the complication of analytical methods, and the need for expensive precision equipment to define the concentration of dozens of individual organosulfur impurities to be determined at fractions in ppm <sup>[12-13]</sup>. In many cases, a feedstock model was used in the study of the hydrotreating process in which dibenzothiophene was used as the only one generalizing hydrogenated component instead of a set of organosulfur impurities <sup>[14-17]</sup>. Most often, researchers consider the grouping of organosulfur impurities of the same homologous series as a certain conditional pseudo-component <sup>[18-20]</sup>, but they do not take into account the fact that homologues with significantly different boiling points also have different reactivity. In this regard, we proposed to consider the feedstock of the hydrotreating process as a set of several narrow fractions, in each of which the aggregate of organosulfur impurities is considered as a conditional pseudo-component. Its concentration in a narrow fraction is determined using the simplest and most accessible analysis for total sulfur <sup>[21]</sup>, that made it possible to significantly simplify the generated mathematical models for the comparative analysis of various hydrotreatment schemes.

# 2. Computational procedures and programs

Since the task of comparing different versions of the reactor units of the hydrotreatment process from the point of view of removing organosulfur impurities was resolved in this work, there was no need to form and use a complex hierarchical model of catalytic hydrotreatment. In this regard, the operation of the reactor was examined on the basis of the following assumptions:

- quasi-homogeneous reaction stream within the reactor;
- constant velocities of local flow jets and hydrodynamic mode of ideal displacement in the reactor;
- isothermicity of the process;
- stationary nature of the process;
- constant activity of the catalyst;
- the content of total organosulfur in the feedstock or in its narrow fractions is considered as an organosulfur pseudo-component and its concentration in the stream is determined by analysis for total sulfur;
- the rate constants of the hydrodesulfurization reactions of the pseudo-components are effective.

Under these assumptions, the mathematical model of a hydrotreating reactor for N pseudocomponents takes the following form of a system of equations:

$$\frac{dC_{S1}}{d\tau} = -K_1 C_{S1N}$$

$$\frac{dC_{S2}}{d\tau} = -K_2 C_{S2}$$

$$\vdots$$

$$\frac{dC_{Si}}{d\tau} = -K_i C_{Si}$$

$$\vdots$$

$$\frac{dC_{SN}}{d\tau} = -K_N C_{SN}$$
(1)

where  $C_{Si}$  and  $K_i$  are the concentration of the organosulfur pseudo-component and the effective reaction rate constant of the i-th pseudo-component, respectively.

The numerical values of the concentration of organosulfur components in the hydrotreating feedstock and the effective rate constants of the hydrodesulfurization reactions were taken from independent literature sources.

The mathematical model was calculated by the Runge-Kutta method and the calculation program made it possible to determine the change in the concentration of pseudo-components during the process, the residence time of the reaction mixture in the reactor until a given degree of purification of feedstock was achieved, and the required volume of catalyst in the reactor. The same assumptions were used when comparing different configurations of reaction units. It is the specific features of the hydrodesulfurization chemistry in these units that made it possible to quickly compare them in terms of the effectiveness of the hydrotreating process using a fairly simple calculation program instead of complex programs that fully take into account the system of hierarchical modeling of the catalytic process from the catalyst grain to the reactor taking into account its placement in the reaction unit scheme.

#### 3. Results and discussion

# **3.1.** Analysis of efficiency of industrial reactor units of diesel fuel hydrotreating plants

The variety of types of layout for the reactor unit of hydrotreating plants (Fig. 1) requires their comparison from the standpoint of minimizing the loading of the catalyst to obtain highquality diesel fuel. Since the reactors are structurally the same in all circuits, when solving the problem, it was assumed that in the reactors, with the assumptions made, the temperature and concentration profiles in the height of the catalyst layer should be practically the same.



Fig. 1. Concepts for reactor units of the process of hydrotreating of diesel fuel a – single-reactor system, b – block double-reactor with parallel the supply of raw materials, c – double-reactor unit with a consistent supply of raw material, d – three-reactor unit with a parallel-to-serial flow of raw materials, d – single-reactor system with recirculation of purified diesel fuel, e – double-reactor unit with differential input preliminary fractionation of raw materials into various reactors.

1 - reactor, 2 - separator, 3 - stabilization column. I – a mixture of raw materials with hydrogen-containing gas, II - blowing off of hydrogen-containing gas, III - light hydrocarbons and hydrogen sulfide, IV - purified diesel fuel. To compare different hydrotreating reactor systems (Figure 1), mathematical modeling of the hydrodesulfurization process of 250 m<sup>3</sup>/h of diesel fuel with a sulfur content of 6000 ppm, the volumetric feed rate of feedstock 2 h<sup>-1</sup> and the accepted effective rate constant of the hydrodesulfurization reaction 2 h<sup>-1</sup> was performed <sup>[12]</sup>. The efficiency of the reaction system in ensuring the final concentration of sulfur in purified diesel fuel from 500 to 10 ppm was estimated by the total volume of the catalyst in the reaction system, represented by a single reactor, two parallel reactors, two sequentially operating reactors, three parallel-sequentially operating reactor (Figure 1, a, b, c,). Calculations have shown (Table 1) that the volume of the catalyst in a single reactor and in a parallel and sequential reactor system required to achieve a given residual sulfur concentration in diesel fuel is the same and the configuration of the installation scheme is determined by technical limitations. This conclusion is valid not only for two-three reactor circuits, but also for a four-reactor <sup>[22]</sup>, as well as for a combined parallel-serial circuit (Figure 1, d), allowing us to assume that all actual industrial reactor units, regardless of their configuration and number of reactors, are equivalent to the operation of a single reactor.

Reaction system	Volume flow rate of recirculate, m <sup>3</sup> /h	Final concen- tration of sul- fur in purified fuel, ppm	Duration of reaction, h	The volume of the catalyst in the system, m <sup>3</sup>
Systems (Figure 1, a, b, c, d) - single-reactor system, block double-reactor with parallel the supply of raw materials, dou- ble-reactor unit with a con- sistent supply of raw material,	0	2000	0.550	68.8
Systems (Figure 1, a, b, c, d)	0	500	1.245	155.6
Systems (Figure 1, a, b, c, d)	0	350	1.420	177.5
Systems (Figure 1, a, b, c, d)	0	50	2.395	299.4
Systems (Figure 1, a, b, c, d)	0	10	3.199	400.0
System (Figure 1, e) - single- reactor system with recircula- tion of purified diesel fuel	10	500	1.220	164.7
System (Figure 1, e)	30	500	1.175	182.1
System (Figure 1, e)	50	500	1.130	197.7
System (Figure 1, e)	10	350	1.400	189.0
System (Figure 1, e)	30	350	1.350	209.2
System (Figure 1, e)	50	350	1.310	229.2
System (Figure 1, e)	10	50	2.370	319.9
System (Figure 1, e)	30	50	2.325	360.4
System (Figure 1, e)	50	50	2.285	399.8
System (Figure 1, e)	10	10	3.175	428.6
System (Figure 1, e)	30	10	3.130	485.1
System (Figure 1, e)	50	10	3.085	539.8

Table 1. Results of mathematical modeling of various reaction blocks of diesel fuel hydrotreating plants

For a single reactor proposed in the patent <sup>[23]</sup> with a purified diesel fraction fed into it as a recycle after separation of hydrogen-containing gas (Figure 1, d), it was assumed that the reaction duration should have been reduced by reducing the sulfur concentration at the reactor inlet. However, mathematical modeling has shown that the supply of a recycle of purified diesel fuel from the separator to the reactor is not rational, since this technique leads to an increase in the reaction volume, all other things being equal (Table.1).

Calculations made it possible to detect the anti-battery effect of the recycle feed on the main parameters characterizing the hydrotreating process (Figure 2). When the recycle is fed to the top of the reactor, the concentration of sulfur in the mixture of feedstock and the recycle only slightly reduces the required duration of contact of the reaction mixture with the catalyst,

but at the same time the volume of flow increases significantly and, as a result, the required volume of the catalyst in the reactor increases. The supply of the recycle to the zones of the catalyst layer different in height of the reactor also negatively affects the results of hydrotreating diesel fuel.



Fig. 2. The dependence of the reaction duration (line 1) and the volume of the catalyst (line 2) on the recirculate flow rate at a residual sulfur concentration in diesel fuel of 10 ppm

Sulfur concentration $C_s$ in narrow fraction number 7 in product at									
M=4						M=	=8		
Z	C <sub>s</sub> , ppm	Z	C <sub>s</sub> , ppm	Z	C <sub>s</sub> , ppm	Z	C <sub>s</sub> , ppm	Z	C <sub>s</sub> , ppm
5	10-37	9	10-33	13	1.8*10 <sup>-10</sup>	9	10-37	13	10-12
6	10-37	10	10 <sup>-27</sup>	14	6.8*10 <sup>-6</sup>	10	10-30	14	10-6
7	10-37	11	10-21	15	2.3*10 <sup>-2</sup>	11	10-23	15	2.3*10 <sup>-2</sup>
8	10-37	12	10 <sup>-15</sup>	16	119.8	12	10-17	16	79.8
Reaction time 2.46 h						Reaction ti	me 2.	66 h	

Table 2. Composition of products leaving reactor R-2, based on total sulfur content in narrow fractions at different boundaries of division M of the feedstock into wide fractions

For the transition of the degree of hydrotreatment of feedstock from the residual sulfur content of 350 ppm to the level of 10 ppm during the process with the recirculation feed, it is necessary to increase the duration of the process and, accordingly, the catalyst loading into a single reactor in comparison with any of the schemes in Fig. 1, a, b, c and d by 1.2-2.2 times with a recirculation coefficient of 0.1-1.0.

Computer calculations of mathematical models of various versions of reaction units of industrial diesel hydrotreating plants have shown that further improvement of existing plants with the enlargement of the hydrotreating process is possible only through the use of a more active catalyst or an increase in the volume of reactors.

#### 3.2. Modeling and optimization of promising method of hydrodesulfurization of diesel fuel

In the works of Loginov *et al.* that remained unnoticed by the industry <sup>[24-25]</sup>, an original idea was expressed about the possibility of separate hydrogenation of two fractions of diesel fuel (light I' and heavy I") in the reactors of a hydrotreating plant under different operating modes of reactors, taking into account the distribution of easily hydrogenated and difficult-to-

hydrogenate organo-sulfur in light and heavy feedstock (Figure 1,e) and an experiment was performed on a pilot plant for hydrotreating diesel fuel boiling within 180-360°C, divided into two broad fractions 180-300°C and 300-360°C with an arbitrary division boundary of 300°C <sup>[25]</sup>. In the development of this idea, we have substantiated the position that when dividing the feedstock into two separately hydrogenated broad fractions, there is such a temperature division boundary at which the costs of the process catalyst and its quantity can be minimized <sup>[26]</sup>.

As the initial data for mathematical modeling of the separate hydrotreating of diesel fuel and the characteristics of feedstock, we took the results of analysis of the diesel fuel fraction 180-360°C in terms of total sulfur content and experiments on a pilot plant 16500 ppm <sup>[17]</sup> that made it possible to describe the composition of feedstock in terms of sulfur by the following equation:

#### $C_{\rm S} = 5000 + 11500 \text{N}/16$

(2)

where  $C_s$  is the concentration of total sulfur in the narrow fraction, ppm; N is the serial number of the narrow fraction as the boiling point of diesel fuel increases; 16 - the number of narrow 6.25% v/v fractions in diesel fuel.

The data on the total sulfur content in the catalysts at different times of hydrotreating the diesel fuel fraction of 180-360°C on the CoMo catalyst obtained in <sup>[17]</sup> made it possible to calculate the range of reaction rate constants within 2-16.3  $h^{-1}$ .

For narrow fractions with numbers N equal to 1, 2, 3, ..., 15,16, the values of the reaction rate constants K<sub>i</sub> were taken as 17, 16, 15, ..., 3, 2.



Fig. 3. Schematic diagram of a reactor block with separate hydrogenation of raw materials, previously divided into light and heavy fractions:

1 - rectification column, 2 - reactor R-1 for hydrogenation of light fraction of raw material, 3 - reactor R-2 for hydrogenation of heavy fraction of raw material, 4 - stabilization column At the first stage of modeling the process of diesel fuel hydrotreating with separate hydrogenation of two wide fractions (Figure 3), the influence of the feedstock division boundary in a complex of 16 narrow fractions with two resulting broad fractions for the required amount of catalyst necessary for purification of wide fractions from sulfur up to 10 ppm was considered. The consumption of the feedstock is taken to be 100 m<sup>3</sup>/h, the volumetric feed rate of the feedstock is 1 h<sup>-1</sup>.

Fractions with numbers 1, 2, ..., M were introduced into the first reactor R-1sequentiallyat the fission boundary between the M-th and M+1-th fractions; fractions with numbers M+1, M+2,...N were introduced into the second reactor R-2. The boundary fraction M changed its number from 1 to N, thus all 16 possible variants

for the distribution of the feedstock between the two reactors were calculated. At M = N, all feedstock was introduced into the reactorR-1, at M = 0, all feedstock was introduced into the reactor R-2 - these calculation options corresponded to the operation of the plant with a single reactor or with two parallel reactors or with two reactors operating in series and gave the same results of calculation.

Mathematical modeling of the hydrotreating process made it possible to understand the characteristic features of hydrodesulfurization of organosulfur impurities in two reactors.

The R-1 reactor is characterized by an obvious increase in the required contact and catalyst loading times associated with the weighting of feedstock in R-1 and an increase in its sulfur content as the numbers of narrow fractions entering the reactor R-1 increase.

Similar dependences for the reactor R-2 are somewhat paradoxical, since with the increasing weight of the wide fraction entering R-2, the required catalyst loading decreases (Figure 4),

but they can be explained when taking into account the change in the mass of the diesel fuel flow purified in the reactor. For example, comparing the results of hydrotreating in R-2 on a lightweight wide fraction at M = 4 and a weighted wide fraction at M = 8 (Table 2), it can be noted that when the reactor is operating on both types of incoming feedstock at the outlet of the equipment, the residual concentration of the most difficult-to-hydrogenate pseudo-component of the sixteenth narrow fraction is 10s of ppm, while the specified purity of the catalyzate in terms of total sulfur of 10 ppm is ensured by almost complete removal of sulfur from the lighter narrow fractions.



Fig. 4. Dependence of the volume of catalyst loaded into the reactor block on the number of pseudo-component M at the fission boundary light and heavy fractions for reactors, respectively R-1 (line 1), R-2 (line 2) and R-1+R-2 (line 3)

The combined operation of two reactors of the reaction unit at any value of M is ensured when the catalyst loading is less than that required for the operation of a single reactor and equal to  $231.7 \text{ m}^3$  (Figure 4).

The minimum catalyst loading for a tworeactor hydrotreater is 134.2 m<sup>3</sup> at M = 12 (loading in R-1 is equal to 59.0 m<sup>3</sup> at a wide light fraction flow rate of 75 m<sup>3</sup>/h and that in R-2 is 75.2 m<sup>3</sup> at a wide heavy fraction flow rate of 25 m<sup>3</sup>/h. In addition, it follows from Fig. 4 that when the catalyst loading is slightly higher than the minimum one, the temperature boundary between wide light and heavy fractions can be changed, which does not require a clear fractionation of the feedstock in the rectification column (Fig.3).

Separate hydrogenation of feedstock in two reactors, in comparison with single-flow hydrotreatment, makes it possible to reduce the loading of an expensive catalyst per unit by 42.1% or  $97.5 \text{ m}^3$  (from 231.7 to 134.2 m<sup>3</sup>).

Since calculations have shown that the efficiency of hydrotreating is largely negatively affected by the overloading of the reactor R-1 with heavy narrow fractions, and the reactor R-2 with light narrow fractions (Table 2), there is no doubt that it is expedient to combine a part of these fractions into the third stream of the medium wide fraction subjected to hydrotreating in a separate third reactor R-3. This fraction can be obtained in the distillation column of the hydrotreater plant (Figure 3) as a side stream. The reactor R-3 becomes additional equipment of the unit.

Mathematical modeling of a three-reactor scheme of a hydrotreating unit, all other things being equal, showed that when a set of narrow fractions forming a medium narrow fraction sent to the reactor R-3 is formed, a rather diverse variability of solutions appears on the selection of temperature boundaries for the beginning and end of boiling of the middle fraction. It is characteristic that the transition from a two-reactor to a three-reactor hydrotreating scheme is accompanied by an additional decrease in the total volume of catalyst loading into the reactor unit due to the elimination of the previously discussed factors that negatively affect the operation of the reactors R-1 and R-2 of the two-reactor scheme.

When selecting the temperature limits of the medium wide fraction, the adopted set of narrow fractions provides a monotonic change in the volume of the catalyst loaded into the reactor R-3 (Figure 5), while the calculated volume of the total load in all three reactors also has a minimum (Figure 6), like for a two-reactor scheme. However, if a two-reactor unit has a single minimum of catalyst loading, many local extrema of the minimum type are formed-when calculating a three-reactor unit, and when searching for a global extremum, it is necessary to use a scanning method with a sequential change in the range of narrow fractions that form a wide medium fraction for the new reactorR-3.





Fig. 5. Dependence of the volume of catalyst loaded into the R-3 reactor on the numbers of narrow fractions that form its raw material:

line 1 - narrow fractions 11 ... 14,

line 2 - narrow fractions 12 ... 15,

line 3 - narrow fractions 9 ... 12.

Fig. 6. Dependence of the volume of catalyst loaded into the block on the numbers of narrow fractions, feedstock of the R-3 reactor: line 1 - narrow fractions 11 ... 14, line 2 - narrow fractions 12 ... 15, line 3 - narrow fractions 9 ... 12.

As an example of mathematical modeling of the hydrotreating process in a three-reactor scheme, Figure 7 shows the distribution of total sulfur concentrations by pseudo-components in reactors and the kinetics of the process as a whole.



Fig. 7. Kinetics of hydrodesulfurization of pseudo-components contained in narrow fractions forming reactor feedstock: R-1 (a), R-3 (b) and R-2 (c) (NNF - number of narrow fraction)

An additional feature of the variability of the problems being solved for optimizing a threereactor hydrotreatment scheme is the possibility of not only minimizing the catalyst load in a separate variant of the distribution of narrow fractions of diesel fuel over light, medium and heavy wide fractions, but also selecting such a variant of the simulated scheme, so that at a volume sufficiently close to the global minimum for the total loading of the catalyst, the scheme included similar volumes of catalyst in all three reactors of the scheme, which makes it possible to develop the designed reactors as identical equipment (for example, option 1 in Figure 8).

Considering the required volume of loading an expensive catalyst into the reactor unit as an optimality criterion R, we can assume that each calculation option allows us to determine the local optimum, and their comparison allows us to identify the global optimum for solving the problem (Table 3).

Table 3. Comparison of the characteristics of one-, two- and three-reactor units of plants for differentiated hydrotreatment of diesel fuel

Characteristics of the reactor block	Reactors						
	R-1	R-3	R-2				
One-reactor blo	ck (R= 231.7 m	3)					
Distribution of pseudo-components	116						
Raw material consumption, m <sup>3</sup> /h	100						
The duration of contact of raw materials with							
the catalyst, h	2.31						
Catalyst volume in the reactor, m <sup>3</sup>	231.7						
Two-reactor blo	ck (R= 134.2 m	3)					
Temperature limits of boiling of wide frac-							
tions, °C	180-315		315-360				
Distribution of pseudo-components	112		1316				
Raw material consumption, m <sup>3</sup> /h	75		25				
The duration of contact of raw materials with							
the catalyst, h	0.78		3.00				
Catalyst volume in the reactor, m <sup>3</sup>	59.0		75.2				
Optimal three-reactor block							
(option No. 1 - global o	optimum; R= 1	16.3 m³)					
Temperature limits of boiling of wide frac-							
tions, °C	180-292.5	292.5-337.5	337.5-360				
Distribution of pseudo-components	110	1114	1516				
Raw material consumption, m <sup>3</sup> /h	62.5	25	12.5				
The duration of contact of raw materials with							
the catalyst, h	0.60	1.48	3.35				
Catalyst volume in the reactor, m <sup>3</sup>	37.4	41.9	32.0				
Optimal three	e-reactor block						
(option No. 2 - local c	ptimum; R= 11	.9.4m³)					
Temperature limits of boiling of wide frac-							
tions, °C	180-303.7	303.7-348.7	348.7-360				
Distribution of pseudo-components	111	1215	16				
Raw material consumption, m <sup>3</sup> /h	68.75	25	6.25				
The duration of contact of raw materials with							
the catalyst, h	0.68	2.00	3.70				
Catalyst volume in the reactor, m <sup>3</sup>	46.6	23.1	49.7				
Optimal three	e-reactor block						
(option No. 3 - local c	ptimum; R= 12	3.9m <sup>3</sup> )					
Temperature limits of boiling of wide frac-							
tions, °C	180-270	270-315	315-360				
Distribution of pseudo-components	18	912	1316				
Raw material consumption, m <sup>3</sup> /h	50	25	25				
The duration of contact of raw materials with							
the catalyst, h	0.49	0.97	3.00				
Catalyst volume in the reactor, m <sup>3</sup>	24.4	24.3	75.2				



Fig. 8. Dependence of the volume of the catalyst loaded into the reactors R-1 (1),R-2 (2), R-3 (3), into a three-reactor block (4) and a reactor unit of a typical diesel fuel hydrotreatment industrial unit (5) from the numbers of narrow fractions that form the raw material of the R-3 reactor: option 1 - narrow fractions 11 ... 14, option 2 - narrow fractions 12 ... 15, option 3 - narrow fractions 9 ... 12



Figure 9. Dependence of the total catalyst volume in the reactor block of the diesel fuel hydrotreater on the number of reactors. (The area of local extremums from the condition of the minimum of the optimality criterion R is shaded) It can be assumed that a further increase in the number of hydrotreating reactors to the limit of the number of narrow fractions will lead to a further decrease in the required catalyst load in the reactor unit, but this will most likely be inefficient from an economic standpoint due to an increase in the cost of numerous non-standard equipment (Figure 9). For example, with 16 narrow fractions and their separate hydrogenation in 16 reactors, the reactor loading will vary from 2.3 m<sup>3</sup> in the first reactor to 23.1 m<sup>3</sup> in the sixteenth reactor, totaling 108.5 m<sup>3</sup>, which is only 9.8 m<sup>3</sup> achieved global optimum of 116.3 m<sup>3</sup> (Table 3).

The three-reactor scheme, compared with the two-reactor scheme, makes it possible to reduce the catalyst loading, depending on the option of forming the medium wide fraction, from 7 to 13% and bring the reduction in catalyst loading to 50% from the currently used "one-reactor" variant of diesel fuel hydrotreatment.

A feature of the process scheme of differentiated hydrotreatment is the need for computer control of the process of distribution of wide fractions in reactors with a change in the flow rate and composition of the feedstock.

#### 4. Conclusion

It has been shown by mathematical modeling that the reactor units of industrial diesel fuel hydrotreatment units are equivalent to a single reactor, on the basis of which series or parallel reactor piping schemes with the same total catalyst load are formed. It has been substantiated that in case of differentiated hydrotreatment, when the feedstock is pre-fractionated into two or three wide, separately hydrogenated narrow fractions, it is possible to reduce the loading of an expensive catalyst by 40-50% as compared to traditional schemes of reactor units of industrial hydrotreatment plants.

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To whom correspondence should be addressed: professor Naum A. Samoilov, Department of Petroleum and Chemical Technology, Ufa State Technological Petroleum University, 1, Kosmonavtov St., Ufa, 430062, Russia; <u>e-mail: naum.samoilow@yandex.ru</u>

# Article

# **Open Access**

Discriminating Geological Information by Interwell Connectivity and Heterogeneity

Prince Appiah Owusu<sup>1</sup>, Robert Dery Nagre<sup>2</sup>

<sup>1</sup> Department of Civil Engineering, Kumasi Technical University, Kumasi, Ghana <sup>2</sup> Department of Chemical Engineering Kumasi Technical University, Kumasi, Ghana

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#### Abstract

Analysis was carried out on the reservoir connectivity and its heterogeneity using the flow capacity plots of the producer wells. Lorentz coefficient (Lc) together with the modified Lorentz plots determine the heterogeneity of the reservoir and its connectivity. Flow capacity plots, and Lorentz coefficient are found useful to establish geological features surrounding producer well leading to the identification of heterogeneity orientation and degree of connectivity. Heterogeneity increases with increasing Lorentz coefficient (Lc). Slope of tangent to flow capacity plot gives the type of geological features surrounding producer wells. Orientation flow capacity plots, gives good understanding on how producer wells communicate with the surrounding injectors by either fast flow path, slow flow path or through fractures. In addition, flow capacity plots also suggest percentage influence of injectors on potential pore volume. The paper shows that production performance is affected when the reservoir heterogeneity is near homogeneity. Within this zone, production increases as heterogeneity increases. However, beyond this point, as heterogeneity increases there is sharp decline in production performance. This means that high reservoir heterogeneity has an inverse effect on the production performance of producer wells. Hence, essential methods for permeability improvement are required to enhance production.

Keywords: Heterogeneity; Lorentz coefficient; Well connectivity; Flow paths; Performance.

#### 1. Introduction

Many available reservoirs are heterogeneous in nature with variability in geological situations in and around the neighborhood of injector-producer well pairs. Many approaches have been deployed to infer reservoir connectivity as a lead to delineate the degree of reservoir heterogeneity through performance comparison of production well with the surrounding injectors. Albertoni and Lake <sup>[1]</sup> approximated inter-well connectivity judging from coefficients produced through multiple linear regression. Quantitatively, coefficients in the model show the degree of relationship between wells (producer and the injectors) in a waterflood which can tell the degree of heterogeneity. The idea was furthered by <sup>[2]</sup> to produce understanding of geological characteristics and heterogeneity of reservoirs using well production data for a better decision tool in reservoir development. Some researchers including <sup>[3-7]</sup> explored the use of some geological characteristics such as clay-sand production and erosion, injection pressures and recovery rate to understand the degree of the reservoir flow paths and connectivity (thief zones) in different geological reservoirs and its implications on waterflooding performance. Common in their approach is that reservoirs with thief zones register early water breakthrough leading to uneven sweep efficiency and lower oil recovery because of poor injected water efficiency. These approaches can predict the degree of flow paths; however, they are always time consuming, expensive and results are only suited for near wellbore region.

The idea of connectivity was furthered through the understanding in reservoir storage-flow behaviour which was developed initially as an expression in sweep efficiency of injectors in layered reservoir. This approach dwells on the relativity flow in any layer as a function of pore volume, usually in a flow-storage diagram (Lorenz plots or flow capacity plots). Flow storage diagram ratably suggest the reservoir geology. For instance, when 50 % of flow comes from only 10 % of the pore volume of a reservoir or a layer, it indicates fast flow paths <sup>[8]</sup>.

One sure flow-storage concept is the one developed by Lorenz. The Lorenz plots uses results obtained from reservoir core plug experiments in the form of permeability and porosity, while flow capacity plots are based dynamic data (injection and production). Generally, the said variables can tell a good description of properties variation in the reservoir in and around areas surrounding a producer. Shook <sup>[9]</sup> developed flow-storage diagram based on estimated results from tracer tests. The results show to some degree that the flow capacity plots, based on injection-production data, are likely to trail the flow paths and geological features in a reservoir. The Lorenz plot, suggested by <sup>[8]</sup> is used to form flow capacity plot which will provide a means for discriminating reservoir geological information. The Lorenz curve is a plot of cumulative flow capacity, Fm, versus cumulative thickness, Hm. The aim of the research is to provide an understanding of the physics of reservoir fluid flows using most basic field dynamic data (injection and production data) and use same to establish the communication between injector and producer pairs as means of delineating reservoir heterogeneities, preferential transmissibility trends, and the presence of flow barriers and oil saturations.

#### 2. Materials and methods

#### 2.1. The study area

Zhao Ao oilfield is a sub-basin of Nanxiang basin with an area of 10 km<sup>2</sup> is situated between 111° 00`and 113° 30`E longitude and between 31° 80`and 33° 00`N. Nanxiang is a small, rifted basin developed in the Mesozoic and Cenosoic Cras. This basin is 160 km long and 110 km wide, extending through two provinces of China, with a total area of approximately 17,000 km<sup>2</sup>. The basin is filled with dominantly Paleogene strata, which serve as the main petroleum source and reservoir system. The Nanxiang Basin consists of three uplifts (Shigang Uplift, Sheqi Uplift and Xinye Uplift) and three sags (Nanyang Sag, Biyang Sag and Xiangzao Sag). Historically, the exploration of the Biyang Sag began in 1974. In 1975, the first well was completed, and it confirmed that there are thick layers of resource rocks and multiple layers indicating the presence of petroleum in the Paleogene Hetaoyuan Formation. In 1976, a high-capacity reservoir was found and the Shuanghe Oilfield began to be developed. Zhao Ao with 31 wells, is one of the eight oilfields which were developed after more than 30 years of exploration. Average permeability and porosity for the field in eight single sand layers, are shown in Table 1.

Well type	Well name	Average porosity	Average per- meability	Well type	Well name	Average porosity	Average per- meability
P1	Ann 6	0.15	185.0	P17	Ann 99	0.12	272.5
P2	Ann 7	0.13	260.0	P18	Ann 100	0.13	170.3
P3	Ann 8	0.16	368.0	P19	Ann 101	0.12	84.0
P4	Ann 10	0.14	64.3	I1	Ann 4	0.14	260.0
P5	Ann 39	0.10	65.0	I2	Ann 5	0.15	313.3
P6	Ann 42	0.14	126.7	I3	Ann 25	0.06	11.0
P7	Ann 45	0.14	107.2	I4	Ann 31	0.14	105.0
P8	Ann 49	0.15	120.7	I5	Ann 41	0.13	75.0
P9	Ann 78	0.11	106.2	I6	Ann 44	0.16	245.0
P10	Ann 79	0.08	217.5	I7	Ann 47	0.14	115.6
P11	Ann 86	0.13	192.0	I8	Ann 51	0.14	80.2
P12	Ann 55	0.06	34.0	I9	Ann 57	0.14	173.8
P13	Ann 91	0.11	192.0	I10	B 76	0.16	179.6
P14	Ann 95	0.10	250.0	I11	B 98	0.14	173.8
P15	Ann 96	0.15	164.3	I12	B 70	0.11	221.7
P16	Ann 97	0.13	252.5				

Table 1. Average porosity and permeability values



Figure 1. Maps showing the: (a) Location of the Nanxiang Basin in China. (b) Location of the Zhao Ao Oil field (c) Location of Ann Tent Area. (d) Location and distribution of wells used in the study

#### 2.2. Lorenz model

The Lorenz curve (Fig. 2) is a plot of cumulative flow capacity, Fm, versus cumulative thickness,  $H_{m},$  and computed as shown in Eq. 1 and 2.

$$F_m = \sum_{i=1}^{i=m} k_i h_i / \sum_{i=1}^{i=n} k_i h_i$$
  

$$H_m = \sum_{i=1}^{i=m} h_i / \sum_{i=1}^{i=n} h_i$$



Figure 2. Flow capacity storage

(1) (2)

The concept is that for any n layers in a reservoir, layer permeability are arranged in decreasing order so that m = 1 is the layer with thickness h<sub>1</sub> and the largest permeability  $k_1$  whereas m = n is the layer with thickness  $h_n$  and the smallest permeability  $k_n$ . For a reservoir with m to n layers,  $0 \le F_m \le 1$  and  $0 \le H_m \le 1$  for  $1 \le m \le n$ . Due to the layer ordering, the Lorenz plot monotonically increases from m = 1 to m = n with a monotonically decreasing slope. If the medium is homogeneous, all the permeability values are similar, and the Lorenz plot is indicated by straight line. Increasing levels of heterogeneity are indicated by a departure of the Lorenz plot away from the straight line.

The Lorenz procedure can be modified to include porosity in the calculation <sup>[10-11]</sup>. In place of the cumulative thickness, H<sub>m</sub>, the cumulative storage capacity, C<sub>m</sub>, is used, Eq. 3:  $C_m = \sum_{i=1}^{i=m} \phi_i h_i / \sum_{i=1}^{i=n} \phi_i h_i$  (3)

Like the Lorentz plot, flow capacity plot can be obtained through set of connectivity  $\beta i j's$  and time lag and attenuation  $t_{ij}$  that occurs between stimulus (injection) and production response. The physical interpretation of  $\beta i j's$  of an injector-producer (ij) is given by Eq. 4.  $\hat{q}_i(t) = \beta_{oi} + \sum_{i=1}^{I} \beta_{ij} i_i(t)$   $(j = 1, 2 \dots N)$  (4)

On the other hand, Eq. 5 gives the corresponding time constant  $t = \frac{948 \phi \mu C_t r_t^2}{k}$ 

(5)

(6) (7)

Details of Eq. 4 and 5 are N is the total number of producers and *I* is the total number of injectors. Eq. 4 states that for any given time period, the total production rate of well j  $\hat{q}_j(t)$  is linear combination of the rates of every injector in the field  $\hat{\iota}_i(t)$  plus a constant  $\beta_{oj}$  term. The  $\beta_{oj}$  term is a constant that tries to account for the unbalance in the field.

Similar to Eq. 1 and 3, the new developed flow capacity curve is given as shown in Eq. 6 and 7.

$$F_m = \sum_{i=1}^{i=m} \beta_{ij} / \sum_{i=1}^{i=n} \beta_{ij}$$
$$C_m = \sum_{i=1}^{i=m} \beta_{ij} t_{ij} / \sum_{i=1}^{i=n} \beta_{ij} t_{ij}$$



Figure 3. Different trends of the flow capacity curve in the vicinity of a producer

To illustrate flow capacity and its differential trends with accompanying geological features in the vicinity of the producer, <sup>[12]</sup> presented three major flow trends as shown in Fig. 3. The first curve indicates a secondary porosity (a presence of fractures) in the drainage volume of a producer represented by the steeper segment of the curve; the second indicates that certain injectors communicate with producer through high permeability layers and the other injectors communicate through low permeability layers. For the last curve, the flat behavior shows

that a fraction of the total storage capacity or the total pore volume swept by injectors provides a negligible fraction of the total flow capacity. This is a typical aspect of nonpay zone or a reservoir seal.

### 2.3. Connectivity and heterogeneity

Many studies have been extensively reported to quantify the reservoir complexity (connectivity/heterogeneity) either statically or dynamically.

Connectivity is the fraction of connected reservoir volume (above a permeability/transmissibility threshold) and connected to wells <sup>[13]</sup>. Connectivity can be quantified as nondirectional or directional. Directional connectivity is quantified by connectivity function <sup>[14]</sup>. Connectivity function is similar to semi-variogram function; the connectivity function decreases with the increase of lag distance till it reaches a constant Plateau. Connectivity function derived from continuous properties depends on the property cutoff (such as permeability cut off). Nondirectional connectivity is more commonly used.

McLennan and Deutsch <sup>[15]</sup> used static connectivity parameters which defines the fraction of total connected pore volume. Larue *et al.* <sup>[16]</sup> quantified reservoir connectivity using static connected volume. Connected pore volume directly reflects the well drainage volume as a good indicator of reservoir flow capacity <sup>[15]</sup>.

As a dynamic response, heterogeneity is defined as the dispersity of displacement front of flooding process <sup>[17]</sup>. Statically, heterogeneity is the measure of complexity of flow path and

contrast of permeability. Within geological framework, heterogeneity can either be at core scale, well scale or reservoir scale.

Dykstra-Parson coefficient ( $V_{dp}$ ) and Lorenz coefficient (Lc) are two most used parameters for heterogeneity quantification.  $V_{dp}$  is computed by using quintiles of permeability log-normal distribution. Even though  $V_{dp}$  is robust to log normally assumption, it lacks uniqueness. As such multiple static models could have the same  $V_{dp}$  although, dynamically, they may be different. Again,  $V_{dp}$  has low sensitivity of models to variations in  $V_{dp}$ , when  $V_{dp} < 0.5$  and high sensitivity of models to variation when Vdp > 0.5. Schmalz and Rahme <sup>[8]</sup> introduced Lc, which is defined by the Lorenz plot, a cross plot between flow capacity versus storage capacity. Lc is computed from the area under Lc curve less the area bounded by homogeneity. Lc ranges from 0 to 1 (homogenous to heterogeneous). Like  $V_{dp}$ , Lc is not a unique parameter for characterization of reservoir heterogeneity. However, <sup>[17]</sup> stated that Lc is better than  $V_{dp}$  because it includes porosity or storage capacity and weight.

### 2.4. Discriminating reservoir flow and heterogeneity

Reservoir get heterogenous due to the alteration of composition and structure of rocks by natural geological processes. Using a relative scale of heterogeneity coupled with the unaltered depositional environments, <sup>[18]</sup> and <sup>[19]</sup> have shown that a substantial moveable hydrocarbons get trapped in reservoirs of varying heterogeneity. <sup>[20-24]</sup> evaluated the effects of heterogeneity on hydrocarbon recovery at the bed-scale level. Reservoir heterogeneity is used here to describe the geological complexity of a reservoir and the relationship of that complexity to the flow of fluids through it <sup>[25]</sup>.

Within reservoirs, heterogeneity is assemblages of depositional facies and subfacies; Clastic lithofacies and carbonate lithofacies, with unique characteristics differentiating sediment textures, stratification types, and bedding architectures. Heterogeneity variability is compounded by factors such as post burial alterations of the strata, compaction, cementation, and tectonic deformation. Geological heterogeneities have been classified according to their size or scale, such as wellbore, interwell, and fieldwide scales of heterogeneity (Fig. 4).



Figure 4. Levels of reservoir heterogeneity (modified from Weber <sup>[20]</sup>)

Reservoir property variability at the wellbore scale affect matrix permeability, distribution of residual oil, directional flow of fluids, potential fluid-rock interactions, and formation damage <sup>[26-27]</sup>. Interwell scale heterogeneities affect fluid flow patterns, drainage efficiency of the reservoir, and vertical and lateral sweep efficiency of secondary and tertiary recovery projects <sup>[28-29]</sup>; <sup>[30-33]</sup>. Heterogeneities at the fieldwide scale affect the in-place hydrocarbon volume, areal distribution, and trend of hydrocarbon production <sup>[34-38]</sup>.

Modelling reservoir heterogeneity provides the opportunities for understanding in successful improving performance predictions and interventions in heterogeneous reservoirs.

### 3. Results and discussion

### 3.1. Distribution of reservoir heterogeneity

During the waterflooding period, heterogeneity is an intrinsic factor that determines swept volume and sweep efficiency.



Figure 5. Reservoir heterogeneity

#### jector and production performance of its offsetting producers are the evidence of heterogeneity. The degree of reservoir heterogeneity was therefore evaluated using the Lorentz model as indicated by Eq. 1 and 3. The reservoir categorize into heterogeneous groups and hence its effects on displacement. Fig. 5 shows the heterogeneity distribution of the reservoir with respect to the producer wells. It is obvious that the reservoir heterogeneity is skewed towards reservoir seal trend. The heterogeneity is categorized into three distributions.

To some extent, injection performance of in-

## 3.2. The log-log plot

From Eq. 5, the time for attenuation  $t_{ij}$  between producer *j* and injector *i* is inversely proportional to permeability. Hence the connectivity  $\beta_{ij}$  and the corresponding  $t_{ij}$  are inversely related. For reservoirs with similar properties, each producer communicates with all injectors, a log-log plot of the connectivity and lag time for each producer with the affected injectors should give a straight line with negative slope. Non-homogeneous reservoirs therefore will show a deviation from the straight indicating specific geological conditions in these reservoirs.

The log-log plots of connectivity against lag time for three scenarios; non diffusivity, one month diffusivity and six-month diffusivity conditions indicate three different groups Fig. 6 to 8. Group 1 represents well pairs with large connectivity and low lag time, group 2 represents well pairs with lag time larger than group 1 but with lower connectivity. The last group 3 represents well pairs with the largest lag time with a much lower connectivity. The three groups have cumulative lag time of 42.9 months, 45.73 months, and 80.9 months respectively. Similarly cumulative connectivity for the three groups are 9.81, 4.26 and 7.79. Hence there is cumulative lag time to cumulative connectivity ratio of 4.373, 10.73 and 10.4 respectively.



Figure 6. Log-log plot of connectivity versus lag time for non-diffusivity



Figure 7. Log-log plot of connectivity versus lag time for one month diffusivity



Figure 8. Log-log plot of connectivity versus lag time for six-month diffusivity

## 3.3. Reservoir connectivity and heterogeneity with fractures

Flow capacity plots using the new approach that combines the connectivity and the lag time was developed for the producer and their surrounding injectors. The flow capacity plots for P1, P4 and P12 show a large deviation from 45° line (homogeneous reservoir) Fig. 9.



The flow capacity plots of P1, P4 and P12 indicate two distinct geological conditions in the vicinity of these producers. Two straight lines can be fitted to the flow capacity curve; the first is steeper than the second straight. The steep straight line suggests a large fraction of the total flow capacity is provided by a very small fraction of the total pore volume swept by the surrounding injectors, which is usually an indication of existing fractures in the vicinity of the producers. The second straight line indicates that a little proportion total flow capacity is supported by a large fraction of the total volume of the field; this describes the situation of injectors communicating through the reservoir matrix. The producers in this part of the reservoir have the best of interwell connectivity.

### 3.4. Reservoir connectivity and heterogeneity with sealing trends

The degree of heterogeneity of the reservoir where these wells are located is shown in Fig. 10. The plots show flow capacity plots of each producer.



All flow capacity plots are nonlinear which means that they are heterogeneous in nature. The first straight line parallel to the 45° line, represent a homogeneous reservoir flow capacity with  $L_c = 0$ . Increasing level of heterogeneity is indicated by movement of flow capacity plot away from the  $45^{\circ}$  line with L<sub>c</sub> increasing but less than unity. The producers such as ANN7 (P2), ANN8 (P3), and ANN45 (P7) are completed in six layers, ANN86 (P11) is also completed in five layers, and whiles ANN100 (P18) is completed in four layers. Only ANN79 (P10) is completed in two layers. Each flow capacity plot is made up of two straight lines, which indicate their respective geological feature of the layers the producer is connected to. The first straight line in each plot represents the fraction of total flow capacity provided by the set of injectors surrounding the producers. This straight line with long flat behavior indicates that a fraction of the total storage capacity provides a negligible fraction of the total flow capacity. This is a typical aspect of presence of s reservoir seal. Therefore, all flat straight lines indicate the presence of sealing faults in the vicinity of the producers. The second straight line in all plots suggests that a small fraction of total flow capacity is provided by some fraction of the total volume of the field; this usually contributes from injectors communicating through matrix of the reservoir. The section of the reservoir contained by these well is said to be low reservoir heterogeneity.

#### 3.5. Reservoir connectivity and heterogeneity for different flow paths

The reservoir heterogeneity for this category is shown Fig. 11. The figure shows flow capacity plots for some producers. Producers ANN42 (P6), ANN39 (P5), ANN49 (P8) and ANN78 (P9) are completed in six layers; ANN91 (P13), are completed in four layers, whiles producers ANN96 (P15) is completed in three layers. The flow capacity plots show different geological features in surrounding injectors. Some injectors communicate with the corresponding producer through fast flow paths or high permeability layers and other injectors communicate through slow paths or low permeability layers. This type of heterogeneity of the field indicates that the producers will have significant degree of connectivity. Such heterogeneities at the interwell scale enhances fluid flow patterns, drainage efficiency of the reservoir, and vertical and lateral sweep efficiency of secondary and tertiary recovery projects.









Figure 12. Field heterogeneity distribution

# 3.6. Production performance and heterogeneity

The effect of heterogeneity on production performance is shown in Fig. 13. It is apparent from the Fig. 13 that production performance increases when the reservoir heterogeneity is less than 0.26. Within this region the reservoir behaves more homogeneous, and the well production increases steadily as heterogeneity factor increases.



Figure 13. Effect of heterogeneity on production

# 3.7. Discussion

From the analysis of connectivity and heterogeneity, the pattern of the flow capacity curve is indicative of the geological features present in the surrounding area of a producer well pairs. The reservoir can be said to be divided into three areas. The first case showing a flow capacity

The field wide heterogeneity distribution for both the injector and producers in the reservoir is shown in Fig. 12. From Fig. 12, the injectors with lower degree of heterogeneity are more prone to homogeneous environment. These injectors communicate better with their surrounding producers. However, the dynamic nature of reservoir heterogeneity, the surrounding producers with higher degree of heterogeneity than the injector affects the performance of these reservoirs. There is some degree of backflow or bypass around these producers. This phenomenon makes interwell connectivity more complicated.

The well production performance increases

sharply beyond 0.26 as the reservoir gets

more heterogeneous. However, as heteroge-

neity factor gets bigger, increase in hetero-

geneity lead to a sharp decline in production.

This means that the higher the reservoir heterogeneity there is an inverse telling effect

on the production performance of the pro-

ducers. Hence it is essential that methods to-

ward permeability improvement are consid-

ered to enhance production.

curve which indicates an area of fractures in the drainage volume of a producer represented by the steeper segment of the curve. In the second case, two specific trends are established. The first trend indicates curve showing presence of long flat behavior which shows the fraction of the total storage capacity, or the total pore volume swept by surrounding injectors has negligible effect of the total flow capacity. This is synonymous to nonpay zone or reservoir seal. This means that producers within this part of the reservoir have poor communication with the surrounding injectors leading to poor production performance. The second trend in this case is the straight line in all plots suggesting that a small proportion of total flow capacity is supported by some fraction of the total volume of the field contributed from injectors communicating through matrix of the reservoir.

The flow capacity plots of the third case show how injectors surrounding the producers within this vicinity communicate with producers through fast flow paths and slow flow paths (higher and lower permeability layers). The performance of the producers in terms of their production rate as a function of degree of heterogeneity shows that production performance is affected when the reservoir heterogeneity is near homogeneity. Within this zone production increases as heterogeneity increases though marginal. However, beyond this point, as heterogeneity increases production assumes a near constant value.

This means that the higher the reservoir heterogeneity there is an inverse telling effect on the production performance of the producers. On another hand, the logarithm plot of connectivity coefficients and lag time attenuation, ( $\beta_{ij}$ 's versus  $t_{ij}$ 's) shows the degree of the imposed geology in the reservoir.

#### 4. Conclusion

The flow capacity plots, and Lorentz coefficient were found useful in establishment of the geological features surrounding the producer well leading to the identification of heterogeneity orientation and hence the degree of connectivity. Heterogeneity increases with increasing Lorentz coefficient (Lc). Production performance is affected when the reservoir heterogeneity is near homogeneity. Within this zone production increases as heterogeneity increases. However, beyond this point, as heterogeneity increases there is a sharp decline in production performance. This means that the high reservoir heterogeneity has an inverse effect on the production performance of the producers. Hence, essential methods for permeability improvement should be considered to enhance production.

### Symbols

 $\begin{array}{l} k_i = layered \ permeability, \ \mathsf{m}^2 \\ h_i = layered \ thickness, m \\ m, n, = number \ layers \\ F_m = cumulative \ flow \ capacity \\ H_m = cumulative \ thickness, m \\ \phi_i = layered \ porosity, \% \\ C_m = cumulative \ storage \ capacity \\ \beta_{ij} = connectivity \ coeeficient \\ t_{ij} = time \ lag \ and \ attenuation, months \\ \hat{q}_j = total \ production \ of \ layered \ thickness, \ \mathsf{m}^3/\mathsf{d} \\ \mu = fluid \ viscosity, \mathsf{Cp} \\ C_t = total \ ompressibility, 1/psi \\ r_t = distance \ between \ wells, m \end{array}$ 

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To whom correspondence should be addressed: Dr. Prince Appiah Owusu, Department of Civil Engineering, Kumasi Technical University, Kumasi, Ghhana; <u>e-mail</u>: <u>princeappiahus@gmail.com</u>

# Article

Aeromagnetic Evaluation of Basement Topography and Structures in the Nigerian Sector of the Dahomey Basin and Southwest Region

Michael Oluwaseyi Falufosi<sup>1</sup>, Olawale Olakunle Osinowo<sup>2</sup> and Sikiru A. Amidu<sup>3</sup>

<sup>1</sup> Pan African University Life and Earth Sciences Institute, University of Ibadan, Ibadan, Nigeria

<sup>2</sup> Department of Geology, University of Ibadan, Ibadan, Nigeria

<sup>3</sup> Pan African University Life and Earth Sciences Institute, University of Ibadan, Ibadan, Nigeria

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#### Abstract

The petroleum prospect of the Nigerian sector of the Dahomey Basin is evaluated with high resolution aeromagnetic data. The aim is to delineate the basin topography, deduce the presence of sufficient thickness of sediments and identify the presence of subsurface structures that can form hydrocarbon trapping mechanisms. The total field data was subjected to noise filtering, anomaly transformation and enhancement, with a view of performing geologic discrimination of the subsurface, map rock lineaments and determine basement depths. Radially averaged power spectrum was used to evaluate the noise-signal amplitude-frequency range of the data, which assisted with noise and regional field removal with Butterworth filter. Data processing also include removal of the IGRF field, downward continuation and reduction to pole. Generation of the derivatives, analytic signal, Euler depth solution and lineament maps form the basis of geological and structural mapping, including evaluation of basin architecture. Depth estimates from the total field power spectrum range from less than 1 km to 12 km, while 3D Euler Deconvolution indicates depth to basement range from < 500 m in the northern zone and up to 6000 m in the southern region, with a NE-SW trending relatively shallow region of maximum depth of 1500 m separating the western Dahomey Basin from the Eastern Niger Delta. The presence of up to 3000 m thickness of sediments and presence of fractures whose reactivation can form traps, suggest the potential for untapped resources in the unexplored parts of the Nigerian sector of the Dahomey Basin.

Keywords: Dahomey Basin; Basement topography; Structures; Lineaments; Derivatives.

#### 1. Introduction

The Dahomey Basin is a petroliferous basin and commercial hydrocarbon reserves have been discovered in the Keta (Ghana), Seme (Benin Republic) and Lagos (Nigeria) parts of the basin. The basin was previously abandoned due to the Niger Delta oil boom, but recent renewal of hydrocarbon exploration interests in the basin is due to depletion of the Niger Delta hydro-carbon reserve. Discovery of hydrocarbon deposits in commercial quantities off the coast of Lagos in Aje field (1996) and Ogo field (2013) has further lend credence to continued exploration activities in the basin. Resources of economic importance, which had also been the subject of exploration include limestones, phosphorites, ironstones, kaolinite clay and water <sup>[1-2]</sup>.

The acquisition of high resolution magnetic data by the Federal Government of Nigeria over the entire country have opened up opportunities for comprehensive and advanced studies into the geology and economic potential of the Nigerian geologic terrain. Although, literatures abound on the application potential fields in basement and basin studies of Nigeria, these available data are yet to be fully explored and thus, still potentially valuable for further studies. On the Dahomey Basin area, airborne magnetic data are often processed and interpreted for lineament mapping and basement depths. There are magnetic interpretation works on small parts of the basin, such as <sup>[3-4]</sup>, but this study aim to carry out a basin-wide study of the basement topography and structures, in order to evaluate the hydrocarbon potential of the basin.

#### 2. Location and geology of the study area

The study area is located within longitude 2° 30′ E – 6° 00′ E and latitude 6° 00′ N – 7° 30′ N, in south-western Nigeria. The study area covers the eastern Dahomey Basin, referred to as the Nigerian sector of the Dahomey Basin. The study area stretches eastward into the western area of the Niger Delta Basin. The geologic map of the study area, which was produced from the surface expression of outcrops and sediments and published by the Nigerian Geological Survey Agency (NGSA) is presented in Figure 1. Precambrian Basement Complex rocks occupy the northern part of the study area, while sedimentary rocks occupy the southern parts. The chiefly granitic basement rocks terrain is highly fractured and has been described to be block faulted <sup>[5]</sup>. Basement subsidence during the Early Cretaceous resulted in the deposition of thick sequence of continental sediments <sup>[6]</sup>. The major Cretaceous sedimentary lithologies include sands, shales and limestones. The Tertiary to Recent sediments include sands, shales, limestones, mudstones and claystones <sup>[7-8]</sup>. The Cretaceous sands form outcrops around the basement boundary.



Figure 1. Geologic map of the study area

### 3. Methods

The datasets used include airborne magnetic data acquired from the Nigerian Geological Survey Agency (NGSA) and the ETOPO1 digital terrain models (DTM) acquired from the National Oceanic and Atmospheric Administration (NOAA). The aeromagnetic data was subjected to series of processing and anomaly enhancements using Oasis Montaj software.

### 3.1. The airborne magnetic data

The high resolution aeromagnetic data was acquired from the Nigerian Geological Survey. Data acquisitions were carried out with fixed wing Cessna aircrafts, with mean terrain clear-
ance of 80 m. Measurements were recorded at an interval of 0.1 seconds or less, which corresponds to station spacing of approximately 7 m. Flight lines spacing is 500 m with a trend of 135° (NW-SE), while tie lines spacing is 5000 m, with a trend of 45°. The pre-processed data was provided by the NGSA as a 2D grid of total magnetic intensity (TMI), with constant cell spacing of 100 m. The IGRF 2005 model was used in calculation of declination and inclination, since the data was collected from 2005 and 2009. The geographic coordinate system and WGS 84 reference datum were used during data acquisition and pre-processing. The coordinate system was however transformed to the UTM Zone 31N Cartesian coordinate system prior to processing.

## 3.2. Data analysis and processing

A value of 33,000 nT was added to the TMI data as this was removed initially from the field data during pre-processing. In order to correct for the flight height, the data was downward continued <sup>[9]</sup> to 80 m, which corresponds to the height of measurement aircraft above terrain. Butterworth lowpass filter was then applied, with a cut-off wavelength of 2000 m, which is 4 times the original line spacing of 500 m <sup>[10]</sup> to attenuate noise. Butterworth bandpass filtering was later applied after IGRF removal and reduction to pole, to obtain the residual anomaly data. The filter was implemented with long wavelength cut-off of 100 km and short wavelength cut-off of 5 km, thereby leaving the intermediate fields for further processing and anomaly enhancements.

## 3.2.1. Removal of the IGRF field

The IGRF (International Geomagnetic Reference Field) is used to compute a theoretical magnetic field of the earth. The resulting data from the subtraction of the IGRF field from the total field magnetic data is often referred to as IGRF Residual. The IGRF Residual is sometimes referred to as magnetic anomaly <sup>[9]</sup> and it is due to variation in magnetic intensity of crustal rocks, which is what is important in magnetic surveys. In this work, the acquired total field magnetic data is referred to as the commonly accepted total magnetic intensity (TMI), while the IGRF Residual is referred to as the total magnetic anomaly (TMA). The 2005 IGRF model was used for this work, since most of the data was acquired between 2005 and 2007.

## 3.2.2. Reduction to pole (RTP)

Reduction to pole transformation of the magnetic data was implemented to facilitate interpretations <sup>[10]</sup>. The operation affects both the amplitude and phase of the anomaly and transforms the data to that which would have been obtained at the magnetic pole <sup>[11]</sup>, such that the direction of magnetization and ambient field are both vertical and symmetrical above the source body <sup>[12-13</sup>]. The RTP process <sup>[14]</sup> involves Fourier transform of the data into the frequency domain and multiplication of the result by the RTP operator <sup>[15-18]</sup>.

The standard RTP procedure assumes the direction of the earth magnetic field and crustal magnetization is constant. This may be plausible for small-scale local studies, but not valid for large-scale regional studies <sup>[19]</sup>, in which magnetic inclination and declination changes significantly. In order to mitigate the problem, as the study area spans several degrees of longitude, the differential reduction to pole (DRTP) technique was implemented. The DRTP technique <sup>[19-21]</sup> is a modification of the RTP <sup>[14]</sup> technique and its purpose is to facilitate transformation of magnetic anomalies on a regional scale, spanning several degrees of latitude and longitude.

The RTP operator is an inverse function of inclination, and as inclination approaches zero, the filter becomes unstable <sup>[22-23]</sup>. This situation is prevalent around the equator and regions of low latitudes, within which the study area is located. This causes directional amplification of the anomalies and inherent noise <sup>[16,18,24]</sup>. There are several solutions to problem [25-28], directional filtering <sup>[26]</sup>, but the implemented approach is the simple modification of the DRTP filter <sup>[17-18]</sup>. This involves the use of a modified inclination (I' equals 20°) set greater than the true inclination (-12.7°).

## 3.2.3. Derivatives and analytic signal

Derivative filters were applied to the RTP data for delineation of edges and geological boundaries <sup>[29]</sup>. Horizontal (x and y) and vertical (z) derivatives are very sensitive to the edges of bodies and are thus called edge detectors. The total horizontal derivative is the vector sum of the horizontal (x and y) derivatives and peaks over vertical contacts or forms a ridge, if the source is narrow. The tilt derivative, which is the arctan of the ratio of vertical derivative to the total horizontal derivative was also calculated and it produces patterns that are similar to the first order vertical derivative <sup>[9</sup>]. Analytic signal grid was also generated from the RTP data and the result was used for structural and geologic discriminations. The analytic signal concept as used in this work is the 3D analytical signal amplitude or simply 3D ASA for short <sup>[30]</sup>. The analytic signal in 3-dimensions was given by <sup>[31]</sup>, and it is essentially the vector sum of the horizontal (x and y) and vertical derivatives.

## 3.2.4. Centre for exploration targeting (CET) grid analysis

The CET grid analysis algorithm was developed by the Centre for Exploration Targeting (CET), based at the University of Western Australia. The algorithm, provided as a plugin in Oasis Montaj was used for lineament and edge detection. There are two tools available for lineament and edge detection. These include the Phase Symmetry, which enhances the appearance of linear features and Phase Congruency, which detects discontinuities or edges. Phase symmetry is used in detecting ridges/valleys, while phase congruency is used in detecting edges. Phase symmetry involves identifying axes of symmetry. At the point of symmetry the amplitudes of all frequency components of the data are at maximum, and the axis of symmetry in the 2D data, the data is first broken into 1D profiles and analyzed over multiple orientations at varying scales. Phase congruency depends on the fact that edge features occur at points where local frequency components are maximally in phase. At the edge points, all the component waves are in phase. The resultant solution plots are curves, which are then vectorized into straight lines. Lines less than 5 km in length were rejected.

## 3.2.5. Depth to Basement – Euler deconvolution

The conventional Euler deconvolution method of depth estimation was applied to the data <sup>[32]</sup>, in order to determine the depth and location of the source body. It makes use of the three orthogonal (x, y and z) derivatives of the total magnetic intensity (TMI) data, the value of the regional magnetic field and a structural index (SI). The structural index (SI) depends on suspected geometry of the source body and values of 0 and 1 were used for contact and dyke models respectively <sup>[33-34]</sup>.

The target depth range was 0 to 10,000 metres  $[^{4,35-40]}$ , and attempts were made to use various window size ranging from 2000 to 10,000 m  $[^{10,34,41-42]}$ . In the initial calculations, the maximum percentage depth tolerance (dZ) is set at 25 %. The solutions were windowed (reduced) with variable parameters in order to eliminate spurious values. These include the maximum percentage depth tolerance or uncertainty (dZ), maximum percentage location tolerance or uncertainty (dXY) and maximum/minimum depth. The X-Offset and Y-Offset were also used to window the solutions. These represent the x and y distance of Euler solutions from the centre of the moving window. Solutions located farther than half the value of the solution window size are often not reliable [<sup>10</sup>].

The initial computation was done with a window size of 2000 m, and structural index of 0.0, for contact/fracture model. The solutions were windowed to dZ of 10 %, dXY of 15 %, X-Offset of +/- 1000 m and Y-Offset of +/- 1000 m. Upon scrutinizing the depth solutions with well depths, the solutions were discovered to be generally too shallow. The window size was increased to 4000 m, so that depths up to 8000 m could be accepted with a realistic level of confidence, as depths greater than twice the window size are generally deemed to be unreliable <sup>[34,41]</sup>. The accepted solutions thereafter was carried out with a structural index of 1.0 for dyke model and then windowed to dZ of 10 %, dXY of 15 %, X-Offset of +/- 2000 m and Y-

Offset of +/- 2000 m. The remaining solutions were dot plotted and also gridded. The elevation data used for topographic correction of the depth solutions is the ETOPO1 1-Arc Minute digital terrain model (DTM).

#### 4. Results

Geologic and structural interpretations were initially carried out through qualitative interpretation. This is then followed up with quantitative interpretations, involving lineament mapping and 3D Euler calculation of source depths.

### 4.1. Total field magnetic anomaly distribution

The total magnetic intensity (TMI) values (Figure 2) range from 32000 to 34000 nT. The values generally decrease from north to south and from west to east. The northern half of the study area displays complexity in amplitude variations and has strong imprints of short wavelength anomalies. This area is dominated by basement rocks. The southern half is dominated by long wavelength anomalies and the relatively smooth appearance of this area suggests deeper magnetic sources. Magnetic intensity values are generally higher towards the basement terrain and the anomalies trend in an approximately NE-SW direction.





## 4.2. Reduction to pole at low latitude residual magnetic anomaly distribution

The reduction to pole residual magnetic anomaly values (Figure 3) range from -326 to 517 nT. Anomaly trends range from NW-SE in the western and eastern areas, to a predominantly NE-SW trend in the central area. A basement fracture of regional proportion exists around the centre of the study area. This is designated by a NNE-SSW trending negative anomaly (< -70 nT). The Dahomey Basin in south-west of the map shows anomaly values that decrease from east to west, from > 400 nT to < 20 nT. This, including the increasing smoothening of the anomalies in this direction suggests increasing basin depths. The eastern half of the study area is dominated by two NE-SW trending high positive (> 25 nT) anomalous zones, running from north to south, through the middle and eastern end of the map.



Figure 3. Pole reduced residual magnetic anomaly distribution

## 4.3. Horizontal derivatives of the magnetic data

The x-derivative map (Figure 4) indicates a general north-south trend of basement structures, but exact orientations vary considerably across the study area.



Figure 4. X-derivative of magnetic anomaly

Moving from west to east, the structures trend NE-SW in the western part, NW-SE / NE-SW in the central parts and NW-SE in the eastern part. The elongated features are not well defined in the southern parts but their general north-south trend visible. The y-derivative map (Figure 5) shows the general east-west trend typical of y-derivative maps. However, as the

Nigerian basement terrain have been established to have a general north-south trend, this result is only reliable in the regions where the trends are tilted northward, especially around the north-central areas.



Figure 5. Y-derivative of magnetic anomaly

The total horizontal derivatives (Figure 6) produced results similar to that generated by the x-derivative. However, the total horizontal derivative is characterized with short-wavelength noise and relatively poorer contrast.



Figure 6. Total horizontal derivative of magnetic anomaly

However, the general north-south trend of anomalies are still visible, as well as the alternating nature of the structural trends from west to east. The Dahomey Basin in the southwest of the study area is dominated by basement structures that trends almost north-south. This is different from the obvious NE-SW trend over the Niger Delta Basin in the south-east of the study area.

## 4.4. Vertical (Z) derivative of the magnetic anomaly data

The z-derivatives of the magnetic anomaly data is presented in Figure 7. The z-derivative is similar to the x-derivative, although the structures are relatively less continuous. However, they are well defined, with high anomaly contrasts. The predominantly north-south trending derivative anomalies are well defined in almost the entire part of the study area, although in the north, the terrain shows relatively better contrast. The structural is also observed to vary from west to east, with the north-western areas showing a predominantly NE-SW trend. In the basement terrain, the structural trends vary from west-east, from a predominantly NW-SE direction within the western parts, to a predominantly NE-SW trend in the central areas. The structural trend in the eastern parts is predominantly NW-SE, and the anomalies can be observed to extend from the sedimentary terrain in the south-east to the basement terrain in the north. The Dahomey Basin, located in the south-western part of the study area can be separated from the Niger Delta area by its conspicuous north-south trending anomalies.



Figure 7. Z-derivative of magnetic anomaly data

## 4.5. Tilt derivative of the magnetic anomaly data

The tilt derivatives map of the study area, extracted from the magnetic anomaly data is presented in Figure 8. The tilt derivative produces similar results as the vertical derivative, with the major difference being that in the tilt derivative map, the anomalies are narrower. As a result, the peaks are better placed over the subsurface discontinuities, thereby facilitating a relatively more accurate mapping. The general north-south trend of the basement rocks and the changing orientations from west to east are vividly visible. Several fractures of significant proportions can be traced on the map. A NE-SW trend negative anomaly trace is observed to run through the middle of the map. This corresponds to the well-known regional Ifewara-Zungeru fracture <sup>[43]</sup>. Several other fractures of varying proportions are observed in the northwestern and northeastern parts of the study area. The fractures generally trend in the NW-SE direction.



Figure 8. Tilt derivative of magnetic anomaly data

## 4.6. Analytic signal of the magnetic anomaly data

The analytic signal map generated from the magnetic anomaly data is presented in Figure 9.



Figure 9. Analytic signal of the magnetic anomaly data

This enables the characterization of the geologic terrain into basement and sedimentary terrains, and also separates the Dahomey Basin from the Niger Delta. High positive (> 0.03 emu) anomaly amplitudes dominate the basement terrain which occur north of the study area. The anomalies terminate in the transition zones. The Dahomey Basin in the south-western region is dominated predominantly by low amplitude (< 0.01 emu) anomaly signatures. However, the area can be conveniently divided to two parts; the western part, dominated by low

amplitude (< 0.0005 emu) anomalies and the eastern part, dominated by anomaly amplitudes ranging from 0.007 - 0.014 emu. A region of high amplitude (> 0.024 emu) anomalies separates the Dahomey Basin from the predominantly low anomaly amplitude (< 0.01) dominated Niger Delta Basin in the south-east end of the study area.

## 4.7. CET lineament analysis of magnetic data

The edge solutions (Figure 10) are best developed in the basement terrain, but poorly developed in the southern parts. The lineaments have a predominant north-south direction, but the earlier identified variation in structural trends is well defined.



Figure 10. CET edge solution overlaid on magnetic data



Figure 11. CET ridge solution overlaid on magnetic data

Traces in the western part of the study area trend mainly NW-SE. The central areas contain predominantly NE-SW trending traces. Lineaments that trend NW-SE dominate the eastern parts of the study area. The ridge solutions (Figure 11) are well developed in most parts of the study area. Here, the north-south trend of the basement structures is well defined, and the changing orientations across the study area are quite obvious. In addition, there are several basement fractures trending in various directions. The earlier interpreted NE-SW trending regional fracture through the middle of the study area is well defined on the CET maps, especially on the ridge model map (Figure 11). The trace along the peak of the negative anomaly (< -50 nT) in the south-east corner, coincide with the area reported to contain the Chain Fracture Zone <sup>[36,38,43]</sup>.

#### 4.8. Basement topography from magnetic data

The plot of depth solutions generated using the contact (SI = 0.0) model (Figure 12) presents depth values of 150 to 2500 m. The values appear to increase from north to south and from the middle of the map to the western and eastern parts. The basement topography grid (Figure 13) displays a wide variation in depths to basement, with the values ranging from 0 to -3500 m. The depth distribution shows a division of the study area into three geologic zones, which include the basement terrain (north), the Dahomey Basin (south-west) and the Niger Delta Basin (south-east). The plot of depth solutions generated using the dyke (SI = 1.0) model (Figure 14) presents depth values of < 500 to > 4500 m. The sedimentary terrain can be divided into two geologic zones, with the south-western part having depths > 2000 m, and the south-eastern side having depths generally greater than 1000 m. The basement topography grid (Figure 15) displays a rugged topography, with depth to basement values ranging from -150 to -5000 m. The depth distributions also show a division of the study area into three geologic zones. These include the basement terrain, the Dahomey Basin and the Niger Delta Basin. The basement terrain is the shallowest part of the study area, with depths < 1000m. The Dahomey Basin is well highlighted and appears to be the deepest part of the study area. Depths to basement here range from -1500 m to -5300 m.



Figure 12. Depth to magnetic basement from magnetic data (SI=0.0)







Figure 14. Depth to basement solutions from total field magnetic data (SI=1.0)



Figure 15. Basement topography from total field magnetic data (SI=1.0)

## 5. Discussion

Distinction of the study area into basement and sedimentary terrains was visible in the distribution of the total magnetic intensity data, in which the basement terrain is dominated by short wavelength anomalies, indicating shallow sources, while the sedimentary terrain is dominated by anomalies of relatively longer wavelengths, indicating deeper sources. The general geologic and structural trend in an approximate north-south direction <sup>[4,44-46]</sup>. Subsurface evidences on the underlying geology in the onshore and near-offshore parts of the eastern Dahomey Basin, shows the basin to be of undulating topography and the thicknesses of the sedimentary deposits vary markedly <sup>[36-37,39]</sup> and exceeds the minimum 3000 m thickness necessary for the formation and preservation of hydrocarbon in southern parts <sup>[4]</sup>.

Data transformation and anomaly enhancements enables mapping of the geologic contacts, structural trends and delineation of the Dahomey Basin and Niger Delta Basin. The northern parts of the study area, coincides with the basement terrain of south-west Nigeria <sup>[35,47]</sup>, while the southern parts of the study area coincides with the south-west Nigeria sedimentary terrain <sup>[8,47-48]</sup>. A NE-SW trending region of relatively high magnetic analytic signal amplitude (> 0.02 emu) separates the Dahomey Basin from the Niger Delta Basin. The location of this region around middle of the southern part of the study area, corresponds to the part of the study area reported in previous work to contain the basement uplift separating the Dahomey Basin from the Niger Delta Basin, known as the Okitipupa High <sup>[5,38]</sup>.

Interpretations from the derivatives maps suggest that the geologic terrain is highly fractured, with significantly varying structural trends. This justifies the notion that the basement terrain and coastal basins of south-west Nigeria are highly fractured <sup>[39]</sup>, with the basement rocks and overlying sediments bearing imprints of several episodes of tectonism <sup>[3,36,38,49-55]</sup>. In addition to the varying structural trends indicated the presence of several episodes of tectonic events, the Precambrian basement rocks have also been reported to be polycyclic in nature <sup>[56]</sup>, which implies that several distinct rock units occur together and they can be differentiated on the bases of differences in age, mode of occurrences and tectonic records. Lineament extraction using CET edge and ridge analyses add additional credence to the highly fractured nature of the underlying basement rocks. Regional fractures trending NE-SW were mapped in addition to several other small scale fractures, generally trending approximately north-south. In addition to fractures, the basement structures also consist of ridges and the interpreted NE-SW trending Okitipupa High (or Ridge), is the most discussed structural feature of the Nigerian sector of the Dahomey Basin <sup>[5,38,57]</sup>. The Okitipupa Ridge has also been described as the most important basement structure in the Nigerian sector of Dahomey Basin and it affects the structural styles of the overlying sediments <sup>[35]</sup>.

The second result of Euler depth estimation with the contact model (SI = 0.0) differs in some ways from the first result of Euler depth estimation with the dyke model (SI = 1.0). Validation of the basement topography grids with depth information from boreholes drilled to basement reveals that the two solutions work differently for different parts of the study area. Basement depths from the contact model produces relatively shallower depths for the basement terrain, north of the study area, and the results obtained for the Dahomey Basin, southwest of the study area agrees with the borehole depths. However, the contact model produce basement depths that are shallower than the borehole depths, making the depth to basement values obtained unreliable. Basement depths from the dyke model produce depth values that agree more closely with the borehole depth, thereby making the result more reasonable. The dyke model however, produces depth values that are deeper than they should for the basement terrain and the Dahomey Basin.

It can be deduced from the basement topography maps that the basement terrain has an undulating topography. Depths to basement appear to increase from north to south, with the southwestern part appearing to be the deepest part of the study area. Depths to basement appears to increase to an initial value of 500 to 1000 m in the central parts of the study area, before increasing steeply to more than 3000 m in the Dahomey Basin, southwest of the study area. This suggests a step-wise subsidence of the crustal rocks, thereby supporting the notion that the Dahomey Basin is a sag basin <sup>[4]</sup>. In addition, the existence of basement rocks at more than 3000 m below the surface, also indicates that the Dahomey Basin contain the minimum 3000 m thickness of sediments deposits necessary for hydrocarbon formation. The presence of sufficient thickness of sediments for hydrocarbon formation has also been reported by several workers <sup>[40,58]</sup>. Traps within the Dahomey Basin are both structural and stratigraphic <sup>[36]</sup> and reactivation of the basement fractures has the potential to initiate formation of new trapping systems that can facilitate accumulation of hydrocarbon.

## 6. Conclusion

The basement terrain contains anomalies of relatively short wavelengths and high amplitudes, due to shallow source depths, while the sedimentary terrain exhibit smooth gradients. The additional distinction of the south-western Dahomey Basin from the south-eastern Niger Delta was made possible with the derivatives and analytic signal maps. Lineament analyses reveal that the subsurface basement is highly fractured, with the structures trending in varying directions. The fractures could have acted as hydrocarbon migration paths, considering the abundance of tar sands and oil-seeps around the basement/sedimentary transition zones, which has sediment depths of less than 2000 m. The presence of sediment thicknesses of up to 3000 m in the southern ends of the study area, suggests that there are untapped hydrocarbon deposits that could yet be discovered.

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To whom correspondence should be addressed: Dr. Michael Oluwaseyi Falufosi, Pan African University Life and Earth Sciences Institute, University of Ibadan, Ibadan, Nigeria, E-mail: michael.falufosi@gmail.com

# Article

Kerogen Characterization, Organic Thermal Maturation and Source Rock Potential of Paleogene Strata in Bende-Umuahia Area, Niger Delta Basin, Southeastern Nigeria

## Okechukwu N. Ikegwuonu

Department of Geology, Chukwuemeka Odumegwu Ojukwu University, Uli, Anambra State, Nigeria

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#### Abstract

Sediments of Paleogene age in up-dip series of Niger Delta Basin, around Bende-Umuahia axis, where approximately 260 m thick sequences were exposed, have been systematically logged from base to top. This paper aims at evaluating the petroleum source rock potential and organic thermal maturation status of the sediments. Seventeen (17) selected outcrop samples of shale and mudstone were subjected to palynological sample processing using conventional method of acid demineralization for recovering acid-free unoxidized organic residue (kerogen) from sediments. The formations encountered in the study area include, Imo, Ameki, and Ogwashi Formations. Results obtained generally reveal three (3) main palynofacies groups (A, B, and C), based on significant change in the particulate organic matter constituents of the examined kerogen samples. Palynofacies A is mainly characterized by abundant Amorphous Organic Matter (AOM) and frequent opaque debris followed by phytoclasts and palynomorph, while Palynofacies B constituted mostly of abundant opaque debris and frequent phytoclasts followed by AOM and palynomorph, whereas Palynofacies C is dominated by abundant AOM and frequent phytoclasts followed by opaques and palynomorphs. Kerogen type II/III with oil-gas-prone material is suggested for the Imo Formation, having immature thermal maturation status, with spores/ pollen exine color ranging from colourless - pale yellow, while Kerogen type III with gas-prone material is suggested for the Ameki Formation, with immature - slightly mature thermal status, with spores/ pollen exine color ranged from pale yellow - brown. Ogwashi Formation however constituted kerogen type III with gas-prone material, thermally immature with spores/ pollen exine color range of colourless - pale yellow. The kerogen data generated in this work, using transmitted light microscopy, have proven the method as a reliable tool for evaluating petroleum potential in any given sedimentary basins.

Keywords: Palynofacies; Kerogen; Niger Delta Basin; Paleogene; Spores/Pollen grain.

## 1. Introduction

Niger Delta Basin overlies Anambra Basin in the southern parts (Fig. 1), and forms one of the seven sedimentary domains of Nigeria <sup>[1-2]</sup>. The basin is a Cenozoic gross offlap clastic succession built out atop the Anambra Basin and forms part of the western African miogeocline that spread out onto the cooling and subsiding oceanic crust generated as the African and the South American lithospheric plates separated <sup>[3-4]</sup>.

In recent past, palynological study was meant solely for the study of spores and pollen. It now encompasses investigations on all kinds of microscopic organic particles <sup>[5]</sup>, ranging from palynomorphs with well defined morphology, such as dinoflagellates cysts and chitinozoans, to particles of woods and cuticles, structured and unstructured algal remains, and tissues of uncertain origin which are more difficult to categorize <sup>[6]</sup>. Palynofacies and kerogen studies in the Niger delta basin have been widely undertaken by workers, most of which were concentrated on the southern down dip section towards the offshore, due to the availability of data from well logs generated through extensive oil drilling activities in the region <sup>[6-9]</sup>.



Fig. 1. General geological map of Nigeria showing the Cenozoic succession of the Niger Delta Basin, and location and access route of the study area (modified after <sup>[1]</sup>)

In recent times, due to the government policy to discovering more reserves in frontier regions through increased exploration, the quest for oil and gas has shifted to the inland basins. Works on the outcropping up-dip series in northern parts of the Niger delta basin around, Bende, Oba and Onitsha area, have been carried out <sup>[10-16]</sup>. These works centered mostly on palynostratigraphy and paleoenvironmental studies. Very few works have been able to demonstrate the usefulness of palynofacies and kerogen studies, and other geochemical methods, in assessing petroleum source rock potential and organic thermal maturation within the inland sector of the basin <sup>[4,11,17-18]</sup>.

Okeke and Umeji <sup>[18]</sup> studies palynofacies, organic thermal maturation and source rock evaluation of Nanka and Ogwashi Formations exposed at Umunya and Tollgate sections. They remarked that palynofacies constituents of the Nanka and Ogwashi Formations reflect Kerogen

(Type II / III) with Gas/Oil prone, and (Type III) kerogen with Gas prone, respectively, whereas the thermal alteration index and vitrinite reflectance index (% Ro) values of the formations are consistent with each other. Oboh-Ikuenobe *et al.* <sup>[11]</sup> carried out lithofacies, palynofacies and sequence stratigraphy of the Paleogene strata in southeastern Nigeria, and concluded that the kerogen organic contents of the Imo, Ameki, and Ogwashi Formations are mainly constituted of Type III Gas prone dominated by structured phytoclasts, and with few Type II Oil prone material of marine origin. Nzekwe and Okoro <sup>[17]</sup> investigated hydrocarbon generating potential of the Ameki Formation in Umuahia, southeastern Nigeria using the organic and trace element geochemistry. They noted that the Ameki Formation contains mainly

the inert Type IV kerogen and insignificant gas prone Type III kerogen, with low hydrocarbon generative potential; is immature and was deposited under oxic conditions. Oguanya *et al.* <sup>[4]</sup> examined source rock geochemistry of the Paleogene strata in Bende-Umuahia and environs, using Walkley black wet oxidation and rock-eval pyrolysis techniques. Their result generally reveals a source rock geochemistry of type IV Kerogen with thermal immature status and fair petroleum potential for the formations. This study, therefore, attempts to present a detailed kerogen characterization to evaluation petroleum source rock potential and organic thermal maturation status of Paleogene up-dip outcropping strata in Bende–Umuahia Area, Niger Delta Basin, southeastern Nigeria, using organic-walled microfossils. Different kerogen and palynofacies classification schemes have been proposed by various researchers <sup>[19-23]</sup>. These classification schemes were as result of different preparation techniques and scope of study. The scheme employed here includes Tyson <sup>[21]</sup>, Ibrahim *et al.* <sup>[23]</sup>, and Zobaa *et al.* <sup>[24]</sup> for quick kerogen assessment.

The study area covers approximately 235 km<sup>2</sup> between latitudes of 5°27'N–5°40'N and longitudes of 7°25'E–7°43'E, within the Niger Delta Basin in southeastern Nigeria (Fig. 2). Three Cenozoic formations, including the Imo Formation, the Ameki Formation and the Ogwashi Formation, were encountered in the study area (Figs. 1 and 2).





## 2. Geological setting and stratigraphy

Niger Delta Basin is situated between latitudes of 3°N and 6° N and longitudes of 5°E and 8°E, and occupies the continental margin of the Gulf of Guinea in equatorial West Africa <sup>[1]</sup> (Fig. 1). It has a total area of about 75,000 km<sup>2</sup>, and an entirely clastic sedimentary-fill up to 12,000 m thick. The basin fill was supplied by large continental drainage systems which constructed arcuate and bird-foot deltaic wedges prograde basinward into the oceanic crusts <sup>[25]</sup>. The Imo, Ameki, and Ogwashi Formations have been found to exhibit similar microfloral assemblages with those in the subsurface Niger Delta <sup>[14,16</sup>]. These formations form the up-dip series of the Niger Delta Basin and are the lateral equivalents of the down-dip Akata and Agbada Formations (Fig. 3). The Imo Formation forms the basal stratigraphic unit in the inland part of the Niger Delta Basin. It consists of blue-grey clay and black shale, with bands of

calcareous sandstone, limestone, and marl <sup>[26]</sup>. And, the northward lateral variations into sandstone facies (members) occur in some places such as Ebenebe, Umuna, and Igbabu <sup>[26]</sup> (Fig. 3).



Fig. 3. Stratigraphic synopsis of southern Nigerian sedimentary basins (from given authors)

In western Nigeria, the Imo Formation cut across partially into a thick shaly and in places, arenaceous limestone (the Ewekoro Formation). In the east, the Imo Formation unconformably overlies the Nsukka Formation, exposes at Oduenyi village extending westward through Ndiwo, and terminates at Itu-Mbuzor where it is conformably overlain by the Ameki Formation [16]. The Ameki Formation dominantly consists of alternating shale, sandy shale, clay sandstone, and fine-grained fossiliferous sandstone, with thin bands of limestone [26-28]. It exposed at Bende-Idima road in Itu-Mbuzor town extending northwestward through Ozu-Item to Uzuakoli town and terminates at Amogugu town where it is completely overlain by the Ogwashi Formation [16] (Fig. 3). The Ogwashi Formation consists of alternating coarse-grained conglomeritic sandstone, lignite, and light-colored clays of continental origin [29]. The Benin Formation consists of massive coarse- to fine-grained sandstones and gravels, with minor shale interbeds. The formation is deposited in coastal or alluvial plain setting following a southward shift of the delta into a new depobelt [30].

## 3. Materials and methods

This involves outcrop logging and sampling, and laboratory analysis of collected samples. Exposures in the study area around Bende-Umuahia and environs were systematically sampled from bottom to top at different localities, from Oduenyi village (Loc. 1) in the eastern part (Fig. 2), along Bende-Umuahia road, to Location 35, near Oyivo stream, toward the southwestern end.

Seventeen (17) selected shale and mudstone samples were processed in the laboratory for their kerogen contents, using standard conventional method of acid demineralization for recovering the acid-insoluble organic residue extract (Fig. 4). Each sample was thoroughly cleaned to remove field contaminants. 10 g of each shale and mudstone sample was weighted out and gently crushed with agate mortal and piston. The crushed sample was passed through 36 % conc. hydrochloric acid treatment for 30 minutes to get rid of calcite. The calcite-free sample was digested for 72 hours in 48 % conc. hydrofluoric acid for removal of silicates. The sample was diluted with distil water to neutralize the acid, and then sieve-washed through 10

microns nylon mesh to enable the organic residue be free from mud and acid. The sievewashed 10 g residue equivalent was partitioned into two parts, 5 g each, for oxidation and the other 5 g for kerogen studies.



Fig. 4: Flow chart for palynological laboratory (kerogen) sample processing

Each 5 g kerogen residue extract from each sample was mounted on a glass slide and critically studied using transmitted light microscopy at x10 and x40 magnifications, in order to carry out a qualitative as well as a quantitative analysis of disperse particulate organic matter (POM), determine the palynofacies association and kerogen types, examine spore/pollen exine color, and estimate Thermal Alteration Index (TAI), Vitrinite Reflectance (Ro %), as well as the degree of organic thermal maturation. Each slide was scanned and counted for its (POM) contents, in which the first 200 particles were considered in terms of *rare* (<5 %), *common* (5-15 %), *frequent* (16-35 %), and *abundant* (>35 %) [ $^{31-34}$ ].

#### 4. Results

Unoxidized acid-insoluble organic matter was classified into four main groups similar to those identified by <sup>[21,31,33]</sup>, for quick kerogen assessment. These include:

- Phytoclasts, refer to all structured and dispersed clay to fines and size particles of plant derived kerogen.

- Opaques, defined as all structured dark or carbonized particles of plant-derived kerogen.
- Amorphous organic matter (AOM), refers to all structureless particles of plant-or marinederived kerogen.
- Palynomorphs, refer to all HF acid resistant organic-walled plants remains.

A total of 17 kerogen samples from Locations 2, 8 (Imo Formation), 16, 22, 23, 25 (Ameki Formation) and 33 (Ogwashi Formation) (Fig. 2) were examined for their petroleum source rock potential.

## **Imo Formation**

Table 1 shows the percentage frequency distribution of the total POM present in the samples. From the illustration given in the histogram (Fig. 5), it is evident that organic constituents of the kerogen samples L2/01, L2/02, L2/03, L2/04 from black shale and L8/01, L8/02 from the blue-grey shale are mostly dominated by amorphous organic matter (AOM) and opaque debris, followed by phytoclasts and then palynomorphs.

Table 1. Percentage frequency distribution of the total particulate organic matter (POM) present in samples from the Imo Formation

Particulate organic matter	L2/01	L2/02	L2/03	L2/04	L8/01	L8/02	L8/04	L8/05	L8/06
Palynomorphs									
Spores & pollen	5	2	7.5	6	1.5	2	4.5	2.5	12.5
Dinoflagellate cyst	1.5	3	-	-	17.5	3.5	-	1.5	5
Forams test lining	-	-	0.5	-	1	-	0.5	-	0.5
Phytoclast (structured)									
Plant tissues:									
Tracheids	4	3.5	10.5	10	0.5	1.5	6.5	20	15
Cuticles	-	0.5	-	0.5	1	-	2.5	1	1
Phytoclast (structured)									
Degraded wood	5	6	0.5	1	-	-	2.5	6.5	2.5
Resin	-	-	-	-	-	-	-	-	0.5
Amorphous organic matter									
(AOM)									
Fluffy - Transparent	53	52	16	9.5	45.5	44.5	10	5.5	12.5
Yellow – brown	2	3	2	0.5	3	2.5	1	0.5	2.5
Black brown	-	-	-	-	-	-	-	-	-
Opaque debris	30	30	63	72	30	46	72.5	62.5	48



Fig. 5. Histogram of % frequency distribution of the total Particulate Organic Matter (POM) present in the samples from the Imo Formation



Fig. 6: Micrograph of POM from the black shale at Loc 2, (Imo Formation). (A) opaques; (B) amorphous organic matter; and (C) microfossil (spore)

The AOM was predominantly transparent (coloureless) with few dark fluffy ones. The exine colours of spores and pollen are generally in arrange of colourless to pale yellow. In terms of maturation, the POM was generally immature (Fig. 6). However, samples L8/04, L8/05, L8/06

from the blue-grey shale are dominated by opaques and phytoclasts followed by AOM and palynomorphs. The exine colours of spores and pollen of the samples ranged from colourless to pale yellow and are also immature.

#### Ameki Formation

Table 2 presents the percentage frequency distribution of the total POM present in the samples. The illustration given in the histogram (Fig. 7) shows that organic constituents of the kerogen sample L16 from the calcareous grey shale at the base of Ameki Formation are dominated by amorphous organic matter (AOM) (56 %) and opaque debris (40 %) followed by phytoclasts (3%) and palynomorphs (1 %). The AOM was dominantly transparent (coloureless) with few dark fluffy ones. The exine colours of spores and pollen are generally in a range of colourless to pale yellow. In terms of maturation, the POM is generally immature.

Meanwhile, kerogen sample L22 from the calcareous laminated grey shale at the middle of Ameki Formation is mostly constituted of AOM (42 %) and phytoclasts (33 %) followed by opaque debris and palynomorphs. The exine spores/pollen colouration ranged from colourless to pale yellow and tends to be immature (Fig. 8a).

Table 2. Percentage frequency distribution of the total Particulate Organic Matter (POM) present in samples from the Ameki Formation

Particulate organic matter	L16	L22	L23/01	L23/02	L25/01	L25/02	L25/03	L16	L22
Palynomorphs									
Spores & pollen	1	5.5	0.5	0.5	3.5	4.5	3	1	5.5
Dinoflagellate cyst	-	2.5	-	0.5	1.5	0.5	-	-	2.5
Forams test lining	-	2.5 %	-	-	-	-	-	-	2.5 %
Phytoclast (structured) Plant tissues:									
Tracheids	2	24	17.5	31	17.5	15.5	35	2	24
Cuticles	-	2.5	0.5	3	2	5	11	-	2.5
Phytoclast (structured)									
Degraded wood	1	6	5.5	10	2.5	2	5	1	6
Resin	-	0.5	-	-	2	3	3	-	0.5
Amorphous organic matter (AOM)									
Fluffy - Transparent	49	38	12.5	13.5	1.5	1	1.5	49	38
Yellow – brown	7	4	1	0.5	15	9	11.5	7	4
Black brown	-	-	-	1	0.5	1	0.5	-	-
Opaque debris	40	14.5	62.5	40	54	58.5	50	40	14.5



Fig. 7. Histogram of % frequency distribution of the total Particulate Organic Matter (POM) present in the samples from the Ameki Formation

Moreover, the kerogen samples L23/01, L23/02 and L25/01, L25/02, L25/03 from the mudstones towards the top of Ameki Formation are dominated by opaques and phytoclasts followed by AOM and palynomorphs. However, the exine colours of spores and pollen of kerogen samples L23/01, L23/02 ranged from colourless to pale yellow and are immature, while kerogen samples L25/01, L25/02, L25/03 from the fossiliferous mudstone ranged from yellow-brown to brown and tend to be immature - slightly mature (Fig. 8b).



Fig. 8A: Photomicrographs of POM from the calcareous grey shale at Loc 22, (Ameki Formation). (A) Structured phytoclast; (B) opaques; (C) AOM; and (D) foraminifer linings. B: Micrograph of mature POM from the fossiliferous mud stone at Loc 25, (Ameki Formation).

## **Ogwashi Formation**

The histogram (Fig. 9) shows the percentage frequency distribution of the total POM present in the samples. The kerogen sample L33/03 from grey to dark shale of Ogwashi Formation are dominated by phytoclasts (55 %) and opaques (34%), followed by AOM (7 %) and palynomorphs (4 %) (Fig. 9). The exine colours of spores and pollen ranged from colourless to pale yellow and tends to be immature (Fig. 10 and Fig. 11).



Fig. 9. Histogram of % frequency distribution of the total Particulate Organic Matter (POM) present in the samples from the Ogwashi Formation



Fig. 10. Micrograph of POM from grey shale at Loc 33, (Ogwashi Formation). (A) Structured phytoclast; (B) opaques; (C) AOM; (D) degraded wood; and (E) opaque wood

## 5. Interpretation and discussion

## 5.1 Palynofacies association, kerogen type, thermal maturation and source rock potential

Table 3 shows the summary of kerogen analysis, with some interpretation. Residue extracts from kerogen samples were examined to estimate the color change of fossil exine and compare them using the Pearson's color chart (Fig 12). The distribution of different kerogen components throughout the sections from the oldest to the youngest units, show a significant change in the particulate organic matter (POM) (Fig 12). These changes were recognized in the three formations encountered. Three (3) main palynofacies associations were recognized from the 17 kerogen samples, which include palynofacies (A, B and C).

Table 3.	Summarv	of keroaen	analysis.	with	some interpretat	tion
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FORMATION	SAMPLE NO	PALYNOFACIES ASSOCIATION	S/P COLOUR	TAI	VITRINITE REFLECTANCE (Ro%)	THERMAL MATURATION	KEROGEN TYPE	SOURCE ROCK POTENTIAL
OGWASHI FM	L33/03	Abundant phytoclast & Frequent opaques	Pale yellow - yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE III	Gas prone
	L25/03	Abundant opaque & Frequent phytoclast	Yellow brown - brown	2 to 2-	0.4 % - 0.5 %	Immature to slightly mature	TYPE III	Gas prone
	L25/02	Abundant opaque & Frequent phytoclast	Yellow brown - brown	2 to 2+	0.4 % - 0.5 %	Immature to slightly mature	TYPE III	Gas prone
NOI	L25/01	Abundant opaque & Frequent phytoclast	Yellow brown - brown	2 to 2+	0.4 % - 0.5 %	Immature to slightly mature	TYPE III	Gas prone
FORMAT	L23/02	Abundant opaque & Frequent phytoclast	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE III	Gas prone
AMEKI	L23/01	Abundant opaque & Frequent phytoclast	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE III	Gas prone
	L22	Abundant AOM & Frequent phytoclast	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
	L16	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
	L8/06	Abundant opaque & Frequent phytoclast	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE III	Gas prone
	L8/05	Abundant opaque & Frequent phytoclast	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE III	Gas prone
	L8/04	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
RMATION	L8/02	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
IMO FO	L8/01	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
	L2/04	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
	L2/03	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
	L2/02	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone
	L2/01	Abundant AOM & Frequent opaque	Colourless - Pale yellow	1 to 1+	0.2 % - 0.3 %	Immature	TYPE II/III	Oil-Gas prone



Fig. 11. Graphs of percentage frequency distribution of the (POM) in the studied area

*Palynofacies A* constituted mostly the amorphous organic matter (AOM) and opaque debris, followed by phytoclasts and palynomorphs; *palynofacies B* characterized by opaques and phytoclasts followed by AOM and palynomorphs; and *palynofacies C* dominated by AOM and phytoclasts followed by opaques and palynomorphs.

In the Imo Formation, palynofacies A and B were identified. Kerogen samples L2/01, L2/02, L2/03, L2/04, L8/01 and L8/02, fall within the palynofacies A and interpreted as type II/III kerogen with oil gas prone material, while samples L8/04, L8/05 and L8/06 are grouped under palynofacies B and interpreted as type III kerogen with gas prone material. In terms of maturation, they are generally immature, with exine spores/pollen colour ranged of colourless to pale yellow (Fig 12). Meanwhile, in Ameki Formation, palynofacies A, B and C were recognized. Kerogen sample L16 was characterized by palynofacies A while sample L22 falls within the palynofacies C, and both were interpreted as type II/III kerogen with oil gas prone material. Moreover, samples L23/01, L23/02, L25/01, L25/02 and L25/03 from mudstone are grouped under palynofacies B and interpreted as type III kerogen with gas-



Fig. 12. Composite chart of the studied sequence showing litho-sections & sampling points, lithologic description, palynofacies and kerogen type, and thermal maturation status

prone material. Based on maturation, kerogen samples from fossiliferous mudstone at Loc 25 fall in the mature zone, with exine spores/pollen colour range of yellow-brown to brown (Fig. 12). Other kerogen samples from Locations16, 22 and 23 fall within the immature zone, with exine spores/pollen colour range of colourless to pale yellow. In Ogwashi Formation, palyno-facies B was recognized in sample L33/03 from grey to dark shale, and interpreted as type III kerogen with gas-prone material. In terms of maturation, the exine spores/pollen colour ranged from colourless to pale yellow and falls within the immature zone.

#### 6. Conclusions

Kerogen data generated in this study have revealed three (3) main palynofacies associations namely; *palynofacies A* and *palynofacies B*, and *palynofacies C* based on significant change in the constituent organic particle of the examined kerogen samples. *Palynofacies A* constituted mostly the amorphous organic matter (AOM) and opaque debris; *palynofacies B* characterized mostly the opaques and phytoclasts; while *palynofacies C* is dominated by AOM and phytoclasts.

In the Imo Formation, *palynofacies* A and B were identified, with exine color range of colourless – pale yellow and thermally immature; while in Ameki Formation, *palynofacies* A, B and C were recognized, with exine color ranging from colourless – pale yellow at the base to yellowish brown – brown at the top of the section; whereas In Ogwashi Formation, palynofacies B was recognized, with exine color range of colourless – pale yellow, and thermally immature. These indicate a type of organic matter content that is thermally immature to slightly mature but with a potential to generate mainly gas and less oil.

Kerogen analysis using transmitted light microscopic technique has shown some significant level of confidence, as a reliable tool, and a proxy for more expensive geochemical parameters, such as Rock-Eval pyrolysis. Despite its subjectivity, the method is relatively cheap, quick and, surprisingly accurate for evaluating petroleum source rock potential and thermal maturity status of any given sedimentary basin.

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To whom correspondence should be addressed: Okechukwu N. Ikegwuonu, Department of Geology, Chukwuemeka Odumegwu Ojukwu University, Uli, Anambra State, Nigeria; <u>e-mail</u>: <u>okeynick2010@yahoo.com</u>

# Article

## **Open Access**

Effect of Moisture on the Flowability of the Coal Charge

Denis Miroshnichenko1\*, Valerii Mieshchanin2, Valentine Koval2, Serhiy Kravchenko3

<sup>1</sup> National Technical University Kharkiv Polytechnic Institute, 61002, Kirpychova str. 2, Kharkiv, Ukraine

<sup>2</sup> Ukrainian State Research Institute for Carbochemistry, 61023, Vesnina str. 7, Kharkiv, Ukraine

<sup>3</sup> State Institute for Designing Enterprises of Coke Oven and by-Product Plants, 61002, Sumskaya str. 60, Kharkiv, Ukraine

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#### Abstract

The analysis of temporary norms of technological design of concentrators showed that the known relationship between the average diameter of coal in the range from 0.5 to 75.0 mm and the recommended angles of the gutters is logarithmic. It is shown that an increase in humidity and a decrease in the average diameter of coal leads to an increase in the values of the recommended angles of the gutters, and an increase in the degree of coal metamorphism also lead to an increase in the recommended angles of gutters. These factors act independently. Based on the processing of experimental data, the possibility of sticking the surface of the structural sheet located at different angles, using different variants of coal charge, which is characterized by different levels of humidity, particle size distribution with different productivity for the charge. It is shown that when using the technology of ramming coal charges, increasing the moisture content in the charge from 10 to 12%, the content of particles with a size of 0-3 mm from 90 to 94%, as well as increasing the load of conveyors (from 250 to 350 t/h) clearly leads to an increase in the duration of the charge, i.e. reducing the speed of its movement on the structural sheet, until the occurrence of sticking.

Keywords: Coal charge; Moisture; Gutter; Angle of inclination; Sticking.

#### 1. Introduction

It is known that the scheme of preparation of coal for coking affects the quality of blast furnace coke and technical and economic efficiency of all coke production in general <sup>[1-5]</sup>. Currently, the use of technology of ramming of coal charges in the production of blast furnace coke is widely developed, both abroad and in Ukraine <sup>[6-7]</sup>.

However, there are no scientifically sound recommendations regarding the use of materials for lining the gutters of coal preparation tracts, transshipment units and in the coal tower to prevent the suspension of the coal charge, which has high humidity.

In particular, in <sup>[8]</sup> it is noted that due to the low level ( $91\pm1\%$ ) of grinding and high (11.0-11.5%) humidity of the coal charge in the conditions of the coal preparation plant of CChP PJSC "ArcelorMittal Kryvyi Rih" there is a bad flowability of the coal charge along the gutters of charge facilities and the coal tower. It is established that the use of the maximum possible normative angles of inclination of gutters ( $60-65^\circ$ ) does not provide a reliable descent of the charge.

In view of the above, it was advisable to conduct special studies aimed at studying the impact of raw materials (composition of the charge, its humidity and particle size distribution) and technological (type of construction material, productivity of coal conveyors, height and angle of fall of the coal charge on the gutter) factors on the possibility of its "sticking". Based on the obtained results, it will be possible to recommend scientifically sound values of the angles of inclination of the gutters to prevent sticking of the coal charge in the gutters.

It should be noted that in the temporary norms of technological design of concentrators <sup>[9]</sup> the recommended angles of inclination of gutters for coal grades D and G (Table 1), as well as Zh, K and PS (Table 2).

Class (average diameter) mm	Tilt angles, deg	rees <sup>*</sup> , humidity
Class (average diameter), min	before 7 %	after 7 %
0-100 (50.0)	41-45 (43)	45-50 (47.5)
50-100 (75.0)	25-30 (27.5)	25-30 (27.5)
25-50 (37.5)	30-35 (32.5)	35-40 (37.5)
13-25 (19.0)	35-40 (37.5)	40-45 (42.5)
0-25 (12.5)	50-55 (52.5)	55-60 (57.5)
0-13 (6.5)	50-55 (52.5)	55-60 (57.5)
0-6 (3.0)	55-60 (57.5)	60-65 (62.5)
0-1 (0.5)	75-80 (77.5)	80-85 (82.5)

Table 1. Recommended angles of inclination of gutters for coal of marks D and G

\* Interval and average value

Table 2. Recommended angles of inclination of gutters for coal of the Zh brands,K and PS

Class (average diameter), mm	Tilt angles, degrees, humidity							
Class (average diameter), min	before 7 %	after 7 %						
0-100 (50.0)	45-50 (47.5)	50-55 (52.5)						
0-25 (12.5)	50-55 (52.5)	55-60 (57.5)						
0-13 (6.5)	50-55 (52.5)	55-60 (57.5)						
0-1 (0.5)	75-80 (77.5)	80-85 (82.5)						

In Fig. 1 and 2 were shown graphical relationships between the recommended values of the angles of inclination of the gutters and the average diameter of the coal, and in Table 3 mathematical equations that describe them.





Fig. 1 Graphic relationship between the recommended values of the angles of the gutters and the average diameter of the coal grades D and G

Fig. 2 Graphic relationship between the recommended values of the angles of the gutters and the average diameter of the coal grades Zh, K and PS

Table 3. Mathematica	I equations and	statistical	evaluation

No	Grade of coal	W <sup>r</sup> t, %	Type of equation	R <sup>2</sup>
1		<7	$\alpha = -9.164 \ln(d_s) + 70.015$	0.90
2	D, G	>7	$\alpha = -9.686 \ln(d_s) + 75.606$	0.90
3		<7	$\alpha = -6.715 \ln(d_s) + 70.286$	0.92
4	Zn, K, PS	>7	$\alpha = -6.715 \ln(d_s) + 75.286$	0.92

Analyzing the graphical and mathematical logarithmic dependences, we can conclude that:

- 1. Increasing the humidity of coal leads to an increase in the recommended angles of inclination of the gutters;
- 2. Increasing the average diameter of the coal leads to a decrease in the recommended angles of inclination of the gutters;
- 3. Increasing the degree of metamorphism leads to an increase in the recommended angles of the gutters. The influence of each of the factors is almost independent of the action of others.

## 2. Methods and materials

## 2.1. Raw materials

To determine the influence of raw materials and technological factors that affect the speed of coal charges of real warehouses in the gutters of the coal preparation plant in the case of ramming technology were selected 2 variants of coal charges (Table 4), differing:

- variant 1 - high coal content of the gas group (55%);

- variant 2 - high coal content of brands KS and KP (35%).

Table 4. Component compositions of coal charges

Grade	Participation in the charge,%								
Graue	Variant 1	Variant 2							
G	25	20							
GZhP	30	20							
Zh	25	25							
KS	10	20							
KP	10	15							
Total	100	100							

The results of studies of technological properties, petrographic characteristics and particle size distribution of coal concentrates are given in Tables 5–7. In the Table 5 also shows the results of determining the grinding capacity according to Hardgrove. Analyzing the Tables 5-7 data, it can be stated that the selected coal samples correspond to the accepted quality indicators.

Table 5 Technological	nronartias of the	Icon haibuta
Table 5. Technological	properties of the	Studied Coal

Provider	Grade	Prox	imate a %	nalyses,	Plasto indexe	ometric es, mm	Oxidation index, °C	Hardgrove grindability index, un.
		Ad	$S^{d}_{t}$	V <sup>daf</sup>	х	у	∆t	HGI
Taldynsky incision	G	8.1	0.49	37.9	30	10	6	52
Resource	GZhP	7.3	0.36	37.4	35	13	3	57
Shchedrukhinskaya Farm	Zh	8.9	0.70	34.8	30	27	5	58
Barzas Society LLC	KS	9.4	0.27	19.5	24	9	2	69
Berezivska Foundation	KP	9.4	0.34	24.6	31	10	6	71

 Table 6. Petrographic characteristics of the studied coal

		Petrographic content,						Stages of metamorphism of vitrinite, %					
							RR*	0.50- 0.79	0.80- 0.89	0.90- 1 19	1.20- 1.49	1.50- 1.69	1.70- 2 59
Provider Grade %		%		%	Coal grades, conditionally corresponding to the stages of metamorphism of vitrinite								
		Vt	Sv	Ι	L	$\Sigma FC$	R <sub>0</sub>	DG+G	GZhP+GZh	Zh	К	PS	Р
Taldynsky incision	G	91	0	7	2	7	0.64	100	0	0	0	0	0
Resource	GZhP	75	1	23	1	24	0.68	100	0	0	0	0	0
Shchedrukhinskaya Farm	Zh	88	0	10	2	10	0.95	0	20	80	0	0	0
Barzas Society LLC	KS	24	1	75	0	76	1.19	0	0	52	48	0	0
Berezivska Foundation	KP	39	1	59	1	60	1.05	0	10	84	6	0	0

\*RR-The average arbitrary rate of reflection of vitrinite

Provider	Grade			Partic	le size	distribu	ition (m	ım), %			Average particle diameter, mm
		>50	50- 25	13- 25	6- 13	3-6	1-3	0.5- 1	<0.5	<3	ds
Taldynsky incision	G	0.0	0.0	3.2	12.6	20.4	24.0	12.8	27.0	63.9	3.36
Resource	GZhP	0.0	7.3	15.1	20.5	17.0	15.5	7.0	17.6	40.1	8.73
Shchedrukhinskaya Farm	Zh	0.0	3.5	9.7	18.6	20.4	17.6	8.5	21.7	47.8	6.31
Barzas Society LLC	KS	5.2	12.2	9.6	13.2	12.6	13.7	6.4	27.2	47.2	12.24
Berezivska Foundation	KP	4.6	14.0	6.7	12.9	12.2	13.9	7.8	28.0	49.6	11.90

#### Table 7. Particle size distribution of the studied coal

Indicators of technological properties and petrographic characteristics of coal charge, compiled in accordance with Table 4 are given in Tables 8 and 9. Due to the increased content of gas coal in variant 1, the charge of this variant has a higher value of the volatile matter (33.8%) compared to the charge of variant 2 (31.3%).

Table 8. Technological properties of coal charges

Variant	Prox	imate analys	ses, %	Plastometric in- dexes, mm				
	Ad	S <sup>d</sup> t	V <sup>daf</sup>	х	У			
1	8.3	0.47	33.8	24	14			
2	8.6	0.45	31.3	22	14			

Table 9. Petrographic characteristics of coal charges

				te, %									
			0.50-	0.80-	0.90-	1.20-	1.50-	1.70-					
	Potr	oora	araphic contont %		nt %	RR*,	0.79	0.89	1.19	1.49	1.69	2.59	
Variant	i cu	ogra	Sinc c	oncer	10, 70	%	Coal grades, conditionally corresponding to the stages of metamorphism of vitrinite						
	Vt	Sv	Ι	Г	∑FC	Ro	DG+G	GZhP+GZh	Zh	К	PS	Р	
1	74	0	25	1	25	0.83	55	6	34	5	0	0	
2	65	1	32	1	33	0.90	40	7	43	10	0	0	

\* average arbitrary rate of reflection of vitrinite

## 2.2. Methods

The charge was prepared according to the CC scheme (crushing the charge) to the required size. The coal charge was dumped from a height of ~0.5 (1.5) m on a sheet made of Steel 3, at a distance of ~0.5 m from its top. The moment of collision of the charge with the metal surface and the moment of collision of the charge with the floor surface were recorded. The duration ( $\tau$ ) of the charge movement on the metal sheet was calculated, sec.

- In the framework of the study, the following indicators varied:
- 1. The humidity of the coal charge ranged from 10 to 12%;
- 2. The content of class 0–3 mm varied from 90 to 94%;
- 3. Productivity of conveyors fluctuated from 250 to 350 t/h;
- 4. The angle of the gutter made of Steel 3 varied from 65 to 80°.

## 3. Results and discussion

Table 10 shows the data showing the possibility of "sticking" (+) or "not sticking" (-) the surface of the structural sheet located at different angles, using different variants of the coal charge, which is characterized by different levels of humidity, particle size distribution, with different load.

	W <sup>r</sup> t=10%			W <sup>r</sup> t=11%				W <sup>r</sup> t=12 %				
Variant	$\alpha = 65^{\circ}$	$\begin{array}{c} \alpha \\ = 70^{\circ} \end{array}$	$\alpha = 75^{\circ}$	$\begin{array}{c} \alpha \\ = 80^{\circ} \end{array}$	$\alpha = 65^{\circ}$	$\begin{array}{c} \alpha \\ = 70^{\circ} \end{array}$	$\alpha = 75^{\circ}$	$\begin{array}{c} \alpha \\ = 80^{\circ} \end{array}$	$\alpha = 65^{\circ}$	$\alpha = 70^{\circ}$	$\alpha = 75^{\circ}$	$\begin{array}{c} \alpha \\ = 80^{\circ} \end{array}$
Load 250 t	Load 250 t/h, grinding 90%											
1	+	-	-	-	+	-	-	-	+	-	-	-
2	-	-	-	-	-	-	-	-	+	-	-	-
Load 350 t	Load 350 t/h, grinding 90%											
1	+	+	-	-	+	-	-	-	+	+	-	-
2	-	-	-	-	-	-	-	-	-	-	-	-
Load 250 t	/h, grin	ding 94	%									
1	+	-	-	-	+	+	-	-	+	+	-	-
2	+	-	-	-	+	+	-	-	+	+	-	-
Load 350 t/h, grinding 94%												
1	+	+	-	-	+	+	-	-	+	+	-	-
2	+	+	-	-	+	+	-	-	+	+	-	-

Table 10. Possibility of "sticking	' of a surface of a	constructional sheet
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According to the data obtained, the following can be stated:

1. At  $W_t=10\%$ , "sticking" of coal is not observed at an angle of 75-80°. At an angle of 70°, "sticking" is observed at a load of 350 t/h for variant 1 with a grinding of 90 and 94%, and for variant 2 with grinding only 94%. At an angle of 65° "sticking" is not observed only for variant 2 coal charge at 90% grinding.

2. At  $W^r_t=11\%$ , "sticking" of coal is not observed at an angle of 75-80°. At an angle of inclination of 70°, "sticking" is observed when grinding 94%. At an angle of 65° "sticking" is not observed only for the variant 2 coal charge at a load of 250-350 t/h and grinding 90%.

3. At  $W_t^r=12\%$ , "sticking" of coal is not observed at an angle of 75-80°. At an angle of 70°, "sticking" is not observed at 90% grinding and 250 t/h load and at 90% grinding and 350 t/h load for variant 2. At an angle of 65° "sticking" is not observed only for variant 2 coal charge at a load of 350 t/h and 90% grinding.

Thus, an angle of 75-80° can be considered safe from the point of view of the absence of "sticking" of any variant of the coal charge, any humidity and grinding.

For clarity of perception of the obtained results, graphical dependences (Fig. 3-5) of the duration of coal charge movement by structural sheet were constructed depending on the angle of its inclination at different humidity levels, class content 0-3 mm, conveyor productivity and height of charge drop on sheet.



a) load 250t/h b) load 350 t/h Fig. 3. Graph of the dependence of  $\tau$  on the angle of the surface (variant 1, the height of the fall of the charge 0.5 m, grinding 90%)

Due to the fact that for the charge of variant 2 for 90% grinding, moisture content, angle of inclination and productivity did not affect the duration of the charge, graphic dependencies in this case were not built.



a) load 250t/h

b) load 350 t/h

Fig. 4. Graph of the dependence of  $\tau$  on the angle of the surface (variant 1, the height of the fall of the charge 0.5 m, grinding 94%)



a) load 250t/h



Fig. 5. Graph of the dependence of  $\tau$  on the angle of the surface (variant 2, the height of the fall of the charge 1.5 m, grinding 94%)

Based on the above dependences, we can say that increasing the moisture content in the charge, the content of particles with a size of 0-3 mm, as well as increasing the load of conveyors clearly leads to increased duration of the charge, i.e. reducing its speed on the structural sheet sticking. Table 11 shows the mathematical dependences showing the influence of various factors on the time of emergence of the coal charge on the structural sheet.

Table 11.	Mathematical	dependencies
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No	Variant	Type of equation
5	1	$\tau = -0.3427 + 0.06125 \cdot W_t^r + 0.00005 \cdot N + 0.00857 \cdot (\Sigma 3 - 0) - 0.00653 \cdot \alpha$
6	2	$\tau = -4.9510 + 0.04125 \cdot W_t^r + 0.00078 \cdot N + 0.05552 \cdot (\Sigma 3 - 0) - 0.00315 \cdot \alpha$

## 4. Conclusions

The influence of various factors on the possibility of sticking coal on the gutters during the transportation of the coal charge in its preparation for coking by ramming was studied. Mathematical and graphical dependences that can be used in the preparation of coal for coking by ramming were developed.

## Symbols

 $W^{r}_{t}$  - water content of coal, as received, %;  $A^{d}$  - ash content of coal in the dry state, %;  $V^{daf}$  - volatile matter in the dry ash-free state, %;  $S_{t}^{d}$  - sulphur of coal in the dry state, %;  $R_{0}$  - mean vitrinite reflection coefficient, %; Vt - vitrinite, %; Sv - semivitrinite, %;

- I inertinite, %;
- L liptinite, %;
- $\Sigma$ FC sum of fusinized components, %;
- y thickness of the plastic layer, mm;
- x plastometric shrinkage, mm;
- $d_s$  average particle diameter, mm;
- $\Delta t$  oxidation index, °C;

HGI – Hardgrove Grindability Index, units;

a - gutter angle, °;

- $\tau$  the duration of the charge movement on the metal sheet, sec;
- N load of coal charge on the belt, t/h;
- $R^2$  the coefficient of determination.

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To whom correspondence should be addressed: professor Denis Miroshnichenko, National Technical University Kharkiv Polytechnic Institute, 61002, Kirpychova str. 2, Kharkiv, Ukraine; <u>e</u>-mail: <u>dvmir79@gmail.com</u>

# Article

Application of Chemical EOR in Viscous, Heavy Crude in Thin Stacked Heterogeneous Reservoirs Using CMG Simulator

S. Bhatkar, L. K. Kshirsagar, V. S. Wadgaonkar, P.Dulakhe, V. Jhamtani

School of Petroleum Engineering, MIT-WPU, Pune, India

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#### Abstract

Recovery of viscous, heavy oil from thin stacked reservoirs with heterogeneity is very difficult from sandstone formations. Thermal enhance oil recovery method fails in recovering incremental oil over water flooding. Because of highly viscous crude oil, it becomes difficult to carry out water flooding successfully. Water flooding leads to fingering effect and effective sweep is unable to achieve. Chemical flooding proves to be successful in bringing oil adhere to rock into the production well bore. Alkalii reacts with acid components present in crude oil to form natural surfactants; it reduces interfacial tension (IFT). Polymer solution facilitates in increasing the sweep efficiency and improving the mobility ratio. Surfactant flooding helps in further lowering the IFT. It also helps in altering wettability from oil-wet rock to water wet. Thus making more oil flow able from the rock surface. Core flood experimental runs were performed in laboratory to understand the effect of polymer flooding (PF) and alkali-surfactant-polymer (ASP) flooding on the recovery of viscous heavy oil at reservoir temperature. In this paper, the modeling for core flooding was performed using CMG simulator. CMG STARS was used for modeling the ASP flood. CMG CMOST was used for history matching and forecasting the oil production for ASP flooding. Chemical EOR method proved to be successful in recovering the viscous, heavy oil.

Keywords: Enhanced oil recovery; Chemical EOR; ASP flooding; CMG STARS; CMG CMOST; Polymer flooding.

#### 1. Introduction

Recovery of oil by natural mean is around 15% during the initial primary recovery phase for conventional reservoirs. With the help of secondary recovery, around 30% of the OOIP (original oil in place) can be recovered using water flooding technique. Oil recovery mainly depends on the reservoir rock properties as well as the property of oil inside the rock. It is very difficult to produce the remaining percentage of oil economically because of low sweep efficiency. During 1960 the oil prices were high and the recovery cost was low which forms the basis for the enhanced oil recovery (EOR) to produce the remaining oil, which is available in the trapped formation <sup>[1-2]</sup>.

Petroleum industry faces tremendous challenges in the recovering heavy oil which is viscous in nature from thin stacked payzones <sup>[3]</sup>. There is a rapid decline in production rate because of heavy viscous crude oil which create the adverse mobility ratio effect in the primary recovery method <sup>[4-6]</sup>. The recovery rate of oil is between 10-20% or it can be less than 20% as the recovery of heavy viscous crude oil is very difficult <sup>[7]</sup>. Thermal recovery methods are used mostly for recovery of viscous heavy crude oil <sup>[8]</sup>. In the reservoirs where there are pay zones which contains highly viscous heavy crude oil, chemical flooding is the best option for increasing the incremental oil recovery effectively by the means of enhanced oil recovery method <sup>[9]</sup>. If the crude oil has 200 cP viscosity and has a specific gravity of less than 20 degree then it is termed as heavy oil <sup>[10]</sup>. If there is a significant difference between water viscosity and crude oil viscosity, water fingering effect arises. Water fingering can be caused
due to water encroachment, it leaves back a large amount of oil and ultimately oil recovery is reduced <sup>[11]</sup>.

With the help of chemical flooding recovery of oil can be more than 47 % as in case of (ASP) alkali-surfactant-polymer flooding initially surfactant-polymer (SP) flooding solution is injected after water is pre flushed <sup>[12]</sup>. After the surfactant polymer solution is injected in the formation in order to improve the performance of oil production and increase the recovery of oil a buffer solution is injected in the reservoir apart from using water flooding <sup>[13-14]</sup>. Water flooding can help to recover an incremental oil of around 10% from the OOIP for heavy oil recovery <sup>[15-16]</sup>. Highly viscous heavy oil present in the payzone, where recovery of oil from water flooding is very limited <sup>[17]</sup>. Potential to produce the heavy oil from such reservoir is very good but because of the ineffectiveness caused by the water flooding there is a need to use other EOR methods. In order to increase the recovery from the reservoir in such cases thermal EOR methods have limitations <sup>[18]</sup>. Need for chemical flooding specially alkali surfactant polymer flooding is well understood in the reserves having thickness less than 10m in thickness <sup>[19]</sup>. If the reservoir rock contains heavy crude oil, thermal recovery methods fail to get the incremental oil gain. From the result it's easy to conclude that along with ASP flooding, even the polymer flooding is very effective at low oil saturation levels in recovering oil <sup>[20]</sup>.

#### **1.1. Reservoir heterogeneity**

Heterogeneity in the reservoir or in the formation, then it gives birth to several problem related to recovery of oil. If there is clay present in the formation and if we use water flooding method then it may lead to swelling of clay. Swelling of clay will create problems related to recovery of oil. Also there might be some problems related to heterogeneity which may lead to poor permeability of the formation, which also may lead to reduced recovery of oil. Now in case when we focus mainly on the recovery of heavy oil, then pay zone formation must have good permeability <sup>[24]</sup>. The pay zone must have zero or minimum heterogeneity for good recovery of oil, as permeability also depends on heterogeneity of the pay zone formation. For heavy oil, where viscosity is very high, permeability of the pay zone must be very good for easily making the viscous crude to flow through the pay zone. If the permeability of pay zone is poor due to heterogeneity, then the recovery of heavy oil won't be very good. In case where recovery of heavy oil is not good due to permeability related problems, then application of EOR methods can be done for increasing the oil recovery. But in case of stacked pay zones with heavy oil, having heterogeneity problems, it becomes difficult to use thermal EOR methods for the recovery of heavy crude. Hence in such cases other EOR methods can be used such as chemical flooding, which includes alkali- surfactant (A-S) or alkali- polymer (A-P) or surfactant- polymer (S-P) or even (ASP) flooding can applied. Now amongst these various chemical flooding methods which will be best suited depends upon the rock and fluid conditions and their characteristics, which varies from case to case.

# 1.2. Thin stacked payzones

Recovery of maximum amount of heavy oil from pay zone is a task for the petroleum companies. Water flooding fails as water breaks through the heavy oil due difference in the density and viscosity <sup>[25]</sup>. Also water flooding becomes ineffective as water is not able push the heavy oil in the production wells. Hence, there comes the need for the tertiary recovery. But tertiary recovery application methods are relatively much costlier than the secondary recovery methods. Important and conventional tertiary methods applied for recovery of viscous oil are SAGD (steam assisted gravity drainage), Steam flooding, Insitu combustion. Even these methods of thermal EOR of tertiary recovery fails to recovery heavy oil with thin stacked reservoirs. Hence there is need to find some alternative methods of tertiary recovery which will not cause problems to thin stacked pay zones. Hence Chemical EOR methods are used in such cases.

# 2. Geological description

Reservoir pay zone thickness of 3-19m with a water contact of 900m, reservoir was initially producing through the assistance of a bottom water drive mechanism wherein the oil was

driven by an active aquifer. Reservoir is dealing with a thin stacked reservoir where the initial pressure was 97.3 Ksc, API gravity of oil being 15°, viscosity of the concerned oil is 270 cP, the pressure and temperature condition at which the first bubble of gas comes out in oil solution is 36.4°. From this moment forth the production rate kept deteriorating, subsequently reservoir was subjugated by waterflooding, that too turned out to be not sufficient economically. Before the well is to be subjected for EOR methods some compatibility screening criteria eventuated and a decision of commencing EOR technique with (ASP) flooding.

Prior to the implementation of ASP on actual field, core flooding analysis was incorporated. On the concerned core, eight core flooding experimental runs were carried out, three ASP flooding and five polymer flooding experimental runs were performed successfully. one successful job was simulated on CMG Stars.

This paper articulates the simulation of a thin stacked reservoir on CMG Stars regarding ASP flooding. It also verbalizes about the consequences of upscaling the results obtained from CMG Stars to the fully functioning field. As well as the author has tried to demonstrate how lab and simulation data plays a vital role in determining the optimal EOR method for any desired field.

#### **3. Experimental procedure**

- a. Prepare core 2 types of core i) original (bariea) ii) synthetic
- b. Dry the core to remove moisture and get dry weight (note: temp of drying and time required)
- c. Put the core in core holder and put molten metal to fix the core
- d. Send it for machining.
- e. Flood the core with (saturate with water) I) ETP water/sea water ii) TWW- tube well water, iii) treated produced water.
- f. Basic data of core like porosity, permeability, saturation, pore volume, dry weight is known.
- g. Displace water with oil with: I) alkali ii) surfactant iii) polymer.
- h. Maintain the hot air oven / core holder temperature at reservoir temp and pressure is to be recorded for displacement.
- i. Final removal of oil / displacement of oil is measured and pore volume (PV) is calculated
- j. After finding the volume then AS volume is found
- k. Polymer volume in terms of displacement is found as per PV
- I. No of days/hrs/ time must be recorded
- m.After collecting the oil/water/displaced fluids then experimentation starts from viscosity, adsorption, polymer trace etc. is carried out.





# 4. Analysis of experiments

This section instantiates few lab results and explains the consequence of polymer & ASP flooding.

a) Polymer flooding

Remaining oil saturation with respect to water flooding and polymer flooding



Figure 2. Remaining oil saturation Sor, after waterflood & polymer flood for all experimental runs

The histogram on the left illustrates after succeeding water flooding in the consecutive 5 Experiments Y axis demonstrates higher values of remaining oil saturation obtained in experiment No.6 i.e., 42.1%. But after conducting polymer flooding amongst the 5 cores, analyst came to know that even after having larger quantity of oil left in the reservoir after water flooding the amount of oil produced from experiment No. 6 is not as efficient as the amount of oil produced from experiment No.1 therefore implying polymer flooding on experiment No.1 instead of 6 is economically as well as practically feasible.



#### 5. Polymer displacement efficiency



Figure 3. Polymer displacement efficiency of all ex- Figure 4. RF & RRF for all experimental runs perimental runs for LCDE over waterflood

Fig.3 illustrates displacement efficiency of polymer over and above waterflooding. Waterflooding job is incorporated with polymer flooding to minimize cost and effectively enhance productivity of oil. Experimental run No. 1 satisfies the above statement since it gives maximum efficiency by the application of both polymer and waterflood.

# 6. RF and RFF comparison for polymer flooding runs

The relationship between RF and RRF of polymer flooding goes hand in hand because as the polymer adsorption increases the mobility ratio decreases. As the polymer adsorption reduces permeability of the core, the above 5 experimental runs justify the effects of polymer adsorption in terms of RF and RRF. In experimental run No. 6 the RF & RRF is significantly less than the other 4 experimental runs. This states that the polymer used in experimental run no.6 is considered to be reliant one compared to the other four as its adsorption properties are less affective to core.

#### b) ASP Flooding



Remaining oil saturation with respect to water flooding and ASP flooding

Figure 5. Remaining oil saturation Sor, after waterflood & ASP flood for all experimental runs

In the Fig.5 remaining oil saturation afte waterflooding and ASP flooding is shown. Remaining oil saturation after waterflooding of experimental run no.4 is 39% while the Sor for ASP flooding is 9% similarly for experimental run no. 7 Sor for waterflooding is 40.8% and for ASP flooding 8.5%. therefore experimental run no.7 for ASP flooding in comparison with waterflooding is more efficient.

# 7. ASP displacement efficiency



Figure 6. ASP Displacement efficiency for LCDE over waterflood & polymer flood

Fig.6 justifies displacement efficiency of ASP flooding over and above waterflooding and Polymer flooding. Experimental run no. 7 shows effective displacement efficiency compared to the other experimental runs since in experimental run no. 7 the Sor after Waterflood was more than that of experimental run no.4 therefore even though experimental run no.4 shows more effective displacement efficiency than Experimental run no. 7 but after considering the abovementioned parameters experimental run No. 7 turns out to be the successful ASP flooded core.

#### 8. Remaining oil saturation for all experimental runs

Since there are 8 cores from which experiment run no. 1,2,3,5,6 was polymer flooded and the rest were ASP flooded. The above fig. illustrates the remaining oil saturation over and above waterflood as well as ASP/ Polymer flood. Experimental run no.1 which incorporates polymer flooding gives higher efficiency i.e., has less remaining oil saturation compared to experimental run no. 2,3,5,6. On the other hand experimental run no.7 has left less remaining oil saturation compared to experimental run no.4 & 8.



Figure 7. Remaining oil saturation for all experimental runs of polymer & ASP flooding





As the fig.8 illustrates the amount of volume injected with respect to the displacement efficiency, for experimental run no.1 the amount of polymer injected is more than the displacement efficiency received (above 60%). While on the other hand the amount of ASP volume injected is kept equal to that of polymer injection (experimental run no.1) with increase in displacement efficiency (above 80%).



Figure 9. cumulative oil production & water cut for ASP flooding.

Fig.9, ASP Flooding experimental run no.7 is segregated into four contiguous results. In the first Part I.e. W/f wherein waterflooding was conducted, watercut was found to be less than oil productivity. This waterflooding formulated a smooth path for ASP flooding, the flooding was proceeded which gave a higher watercut than the water flooding done earlies which is a sign of successful areal displacement efficiency subsequently after receiving a little less water cut during asp flooding polymer buffer was introduced to the core where appreciable oil production rate was obtained with decrease in watercut followed by this chase water was flooded which gave a constant production rate of oil as well as water.Therefore, ASP Flooding experimental run no.7 is simulated on CMG STARS.

# 9. Reservoir Simulation using CMG



Figure 10. Process flow chart of ASP flooding simulation on CMG STARS

As Lab results were commendable for flood job no 07, it was simulated on CMG Stars. In order to forecast reservoir's productivity as a resultant of lab experiment i.e. coreflooding, a model was created on CMG STARS. As shown in Fig.3 there were 3 major steps involved for simulating a reservoir. In 1<sup>st</sup> step the data was generated via lab as well as field, relying on the data given by respective offset wells in the reservoir and lab step 2 was commenced, this

step took place in CMG STARS Builder module wherein a reservoir grid was created on Cartesian lines it was considered as foundation subsequently reservoir components, Rock/Fluid properties, types and number of wells their initial conditions were specified. Following this sequence, the model was out-turned which was proceeded to the contiguous step i.e. step 3. In step 3 the lab and field and the model prepared in CMG STARS was scrutinized in CMOST for history matching. In there, CMOST verified and interpreted the condition of reservoir and determined its forecast productivity.

#### 10. Results and discussion

As CMG STARS verified the data for both cases (lab & field) and compared the efficiency of Waterflood, polymer flood and ASP flood. CMG proposed several Graphs in order to determine a suitable flooding process for the desired reservoir.



Figure 11.Cummulative oil SC and water rate SC, liquid rate SC, GOR SC, cumulative GOR & WOR SC, water mass rate SC vs time. — cumulative oil SC; — water rate SC & Water mass rate SC; — liquid rate SC; — cumulative WOR SC; — cumulative GOR SC

Figure 11. Shows the Cumulative oil produced after implementing ASP flooding on the desired reservoir, it also demonstrate the production of water, liquid and gas simultaneously on Y2 axis. As the process cycle of ASP flood reaches towards the end there is significant increase in cumulative oil production with decrease in water, liquid and gas production is observed.While comparing both the EOR Processes (ASP & Polymer flooding) CMG has yield a graph representing the effective process for reservoir.





Figure 12 illustrates that the application of ASP flooding for this reservoir is most effective and feasible, whereas use of polymer flooding gives less oil production and adds up to the total cost of process. CMG has also verified the mechanism and flow pattern of ASP flooding obtained in lab (Figure 9) by producing a graph of the same trend in STARS i.e. Figure 13.



Figure 13. Cumulative oil SC and water cut vs time using ASP flooding; — green-cum. oil SC, — Blue-water cut SC

The viscous nature of oil caused undesired mobility ratio due to which despite having good permeability and hydrostatic pressure, water cut increased sharply in the early phase of production resulting in low (10%) primary recovery. With the objective to improve recovery, application of EOR was conceptualized. After detailed laboratory investigation on feasibility of chemical EOR processes, ASP was found to be the most feasible chemical EOR technique. ASP flooding is quite effective in thin, stacked payzones with highly viscous heavy oil, where it's difficult to use thermal EOR techniques. Further, as is the common practice pilot project of ASP flooding is recommended in the field for three years followed by polymer buffer for two years.

This work has lead in advancement of knowledge where heavy oil pay zones with heterogeneity creates problems in recovery of heavy oil. Hence, ASP/polymer flooding is applicable to this payzone in the field for recovery of heavy oil from all production wells present in this region. The overall percentage recovery of hydrocarbon was increased from selected chemicals used for ASP flooding. In case of a heavy oil bearing sand-shale reservoir laboratory core studies and simulation studies were carried out for water flood, PF and ASP flood with various combinations. In general, above studies are recommended to decide flood plan for improving recovery of oil. The optimized concentration of fluids in injection water could be decided only after that.

# **11.** Conclusion

This paper anticipated the cumulative production of oil over and above waterflooding, by application of EOR method. It also aimed to determine effective EOR method for thin stacked reservoir. This paper investigated both operations .i.e. polymer as well as ASP flooding, which was evaluated in CMG STARS in coordination with CMOST.

- 1) According to CMG, the cumulative oil to be produced over and above waterflooding for ASP method is 40 % whereas for polymer flooding it was just 15% raise in cumulative oil production after waterflooding.
- 2) The remaining oil saturation after ASP flood was 9% while on the contrary ( $S_{or}$ ) for polymer flooding was found to be 29%.
- 3) ASP flooding had less resistance factor as well as less residual resistance factor compared to polymer flooding.
- 4) The watercut in ASP flooding is significantly less than that of polymer flood.

These results enable us to state that the ASP flooding for thin stacked reservoir has proved to be more effective and will ultimately increase oil recovery in oil wet sandstone reservoir.

Further following recommendations are made for possible future work based on the present study:

- 1. Upscaling of the core flood model to field scale needs to be done using CMG for ASP and Polymer flood, for understanding the effect of total cumulative recovery of oil with more data.
- 2. Based on the recommended doses for ASP flooding, there must be further implementation of the pilot project on field scale for ASP flooding, to understand the benefit of ASP on incremental oil recovery.

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To whom correspondence should be addressed: Dr. S. Bhatkar, School of Petroleum Engineering, MIT-WPU, Pune, India; e-mail: <a href="mailto:siraj.bhatkar@mitwpu.edu.in">siraj.bhatkar@mitwpu.edu.in</a>

# Article

Technical and Economical Evaluation of Using Inline Burners for Reheating of the Process Gas in the Sulfur Recovery Unit

Hamid Reza Mahdipoor\*, Hamid Ganji

Gas Transportation and Processing Technology Development Division, Research Institute of Petroleum Industry, Tehran, Iran

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#### Abstract

The duty of the sulfur recovery unit in oil and gas refineries is to convert  $H_2S$  to elemental sulfur. A proper design and operation of this unit lead to lower  $SO_2$  emissions to the atmosphere. Several configurations can be proposed for a specific acid gas stream and each of them has pros and cons. In this paper, the advantages and disadvantages of using inline burners for reheating the process gas are compared with the conventional reheating method, i.e. the use of steam heat exchangers in terms of technical and economical points of view. The achieved results show that using inline burners will increase the investment costs as well as operational problems and is not recommended to be considered in sulfur plants.

Keywords: Sulfur recovery; Inline burner; Steam heat exchanger; Tail gas treatment.

# 1. Introduction

Crude oils often contain considerable amounts of sulfur compounds which need to be removed as H<sub>2</sub>S gas in the refinery units such as gas oil or naphtha hydrotreatment units. Moreover, raw gas coming from the gas reservoir usually contains non-hydrocarbon species such as H<sub>2</sub>S <sup>[1-4]</sup>. The Sulfur Recovery Unit is applied for converting H<sub>2</sub>S to elemental sulfur in oil and gas refineries. The modified Claus process is the most common method for this aim. As indicated in Figure 1, this process includes a reaction furnace at the beginning of the unit in which about 60% of H<sub>2</sub>S is burned and converted to elemental sulfur at the high temperature of 1000-1400°C, depending on the concentration of the H<sub>2</sub>S in the acid gas stream (EQs.1&2).

$H_2S + \frac{3}{2}O_2 \Rightarrow SO_2 + H_2O$	(1)
$2H_2S + SO_2 \Leftrightarrow \frac{3}{2}S_2 + 2H_2O$	(2)

A waste heat boiler (WHB) exchanged the released combustion heat with water to produce steam (High pressure/Low Pressure/Medium Pressure, depending on the needs of the refinery) as well as cool down the process gas. The temperature of the cooled process gas depends on the pressure and temperature of the saturated steam which is produced in the WHB and usually varies in the range of 200 to 300°C. Therefore, a sulfur condenser may be needed after WHB for more cooling of the process gas and condensing and recovering the produced elemental sulfur in the reaction furnace <sup>[5-10]</sup>.

More recovery of sulfur is achieved in the next catalytic step which involves two or three catalytic converters (reactors) and their down-stream sulfur condensers. Although, equation 2 is an endothermic reaction at high temperatures (above 600°C) and the recovery of sulfur increases with increasing the temperature of the reaction furnace, achieving temperatures as high as the reaction furnace temperature is impossible using the conventional heating methods. Therefore, reaction (2) will be exothermic in the Clause reactors temperature (below  $600^{\circ}$ C). To increase the rate of reaction (2) in the Clause reactors, using the alumina catalyst

is considered which can convert SO<sub>2</sub> to elemental sulfur. Several by-products are produced in the reaction furnace together with elemental sulfur, such as COS and CS<sub>2</sub>. These two compounds can be contributed to a high percent of the pollutants in the tail gas. Therefore in addition to the production of elemental sulfur, COS and CS<sub>2</sub> should be hydrolyzed in the first Claus catalytic reactor at a higher temperature than needed for the equilibrium reaction (2) (about 350°C) by the following endothermic reactions <sup>[8-16]</sup>:

$COS + H_2O \Rightarrow H_2S + CO_2$	
$CS_2 + 2H_2O \Rightarrow 2H_2S + CO_2$	

(3) (4)

Effluent gas from the WHB or sulfur condenser must be reheated to the appropriate temperature for entering the Claus reactors. Generally, the conventional methods for reheating the effluent gas can be divided into two major groups including the direct and indirect methods. In the direct methods, the process gas is mixed with a hot gas stream. For instance, the hot gas bypass method is usually used for reheating the first reactor inlet stream (the hydrolyzing reactor) in which a slip-stream of hot process gases from the WHB is taken and mixed with the first reactor inlet stream. Lower overall sulfur recovery is mentioned as its disadvantage <sup>[12]</sup>.

The other conventional direct method uses inline burners to burn either fuel gas or acid gas to produce a hot gas stream (figure 1-a). This hot gas is then mixed with the reactor inlet streams. In addition to the first reactor, this method can be applied for reheating the inlet streams of the second and third reactors too. Controlling the required combustion air is very critical in this method. Any excess oxygen (above 30 ppmv) can lead to the formation of SO<sub>3</sub> and deactivation of the catalyst or increase the corrosion rate in the downstream equipment. Moreover, a shortage of oxygen can lead to the formation of soot in the case of burning the fuel gas which can plug the catalyst pores and deactivate the catalyst.

On the other hand, the indirect reheat methods use direct-fired heaters or steam heat exchangers to reheat the process gases (Figure 1-b). Electrical reheating can also be used in lower capacities (lower than 100 KW). Although applying steam heat exchangers for reheating the Clause reactor inlet streams is considered an expensive alternative, it may be preferred to prevent the above-mentioned operating problems <sup>[12]</sup>.



Figure1. A typical Claus process with a) inline burners; b) steam heat exchangers

As mentioned above, because of the presence of the unrecovered sulfur compounds such as elemental sulfur, COS, and CS<sub>2</sub>, the overall recovery of sulfur in the modified Claus process is usually limited to 96 to 98 percent, depending on the number of catalytic stages. Therefore, Tail Gas Treatment (TGT) section is applied before the incinerator for achieving more recovery of sulfur and reducing the SO<sub>2</sub> emission under stricter environmental regulations <sup>[17-19]</sup>. SCOT process (Figure 2) is usually selected for tail gas treatment. In this process, at first, all sulfur compounds are converted to  $H_2S$ . Then,  $H_2S$  is selectively absorbed in an Amine contactor and desorbed in the regenerator column (stripper). The released  $H_2S$  is returned to the input of the Claus where it is mixed with Claus acid gas feed. The stream exiting from the top of the absorber column includes a negligible amount of  $H_2S$  and is sent to the incinerator <sup>[18-20]</sup>.



Figure2. Schematic flow diagram of the SCOT process [19]

In the next section, two types of reheating methods, i.e. Case I: using the inline burners (Figure 1-a), and Case II: using the steam heat exchangers (Figure 2-a), are considered in a typical SRU. At first, only the Clause section is simulated to compare these two cases in terms of the technical and economical points of view. Then, the TGT section is added to the simulation and the results are presented for a complete sulfur recovery unit.

#### 2. Case study results

To investigate the effect of the applied reheating method on the performance of SRU, a typical industrial SRU with two catalytic reactors is considered. For this purpose, the commercial simulation software, Promax is applied which is calibrated and verified by several SRU designs and operating data of Iranian refineries to achieve valid and accurate results.

Burning of fuel gas in the inline burners produces  $H_2O$  and  $CO_2$ . According to eq.2, since  $H_2O$  is a product of the Clause equilibrium reaction, increasing the amount of  $H_2O$  is expected to shift the reaction toward the left side and reduce the overall recovery of sulfur. Moreover, increasing the  $CO_2$  gas may have a negative effect on the selective absorption of  $H_2S$  in the absorber column of the TGT section. Therefore, in addition to a Claus section, the considered SRU contains one TGT section to study the influence of applying inline burners as preheating method on the performance of the TGT section, too. The specification of the acid gas stream which is considered as SRU feed is given in Table 1. As indicated in this table, the feed stream is not so lean and the concentration of  $H_2S$  reaches 40-mole percent.

Property	Value
Temperature	60°C
Pressure	1.8 bar
Mole flow	600 kmole/h
Composition (r	nole%)
H₂S	40
CO <sub>2</sub>	46
CH <sub>4</sub>	4
H <sub>2</sub> O	10

Table1. Specifications of the acid gas feed to SRU

High-Pressure Steam (HPS) is considered to be generated in the WHB and therefore a sulfur condenser is needed for cooling the process gas down to  $180^{\circ}$ C and separating the elemental sulfur produced in the reaction furnace. As described before, a high operating temperature of around 250 °C is needed for the first Claus reactor inlet stream to ensure that COS and CS<sub>2</sub> have hydrolyzed appropriately <sup>[12]</sup>. The second condenser is also responsible for the separation of sulfur produced in the first reactor. Unlike the first reactor, a lower operating temperature is needed in the second Claus reactor to enhance the conversion of the exothermic Claus reaction. But a temperature approach should be considered to prevent sulfur condensation in the catalyst bed. Therefore, the second preheater should rise the temperature of effluent gas from the second condenser, from about  $170^{\circ}$ C to  $200^{\circ}$ C.

To investigate the effect of preheating method on the performance of the Claus section, at first, the recycled gas from the TGT section is not considered in the simulations. Table 2 shows the required utility for preheating the inlet streams into the first and second reactors in Cases I and II, i.e. required fuel gas in Case I and required HPS in Case II. In case study II, HPS at 260 °C and 47 bar are used as the heating medium. As presented in this table, the yearly required cost for reheating the process gas in Case I, i.e. using inline burners, is almost similar to the yearly required cost in Case II, i.e. using steam exchangers. However, the initial investment required in Case I will be greater than in Case II due to the need for more complex control systems.

Burning the fuel gas may increase the volumetric flow rate of streams that enter the first and second reactors as well as the tail gas stream which leaves the Claus section toward the TGT section. Increasing the flow rate of reactor and TGT feeds will increase the size of the equipment and, consequently the investment costs. Standard volumetric flow rates of reactor inlet and tail gas streams are presented in Table 2. As shown in this table, the flow rate of the first rector inlet in Case II is 1204 m<sup>3</sup>/h greater than the first rector inlet in Case I (about 4%). This increase for the second reactor inlet and tail gas streams are 1643 m<sup>3</sup>/h (about 5.6%) and 1642 m<sup>3</sup>/h (about 5.7%), respectively. Therefore, in addition to requiring more investment for using inline burners instead of steam heat exchangers, more costs are needed for reactors and condensers in the Claus section. Moreover, it is expected that a larger TGT section is needed too.

As discussed above, in addition to producing  $CO_2$ , burning of fuel gas in the inline burners produces  $H_2O$  which shifts the Claus reaction toward the left side and reduces the overall recovery of sulfur. As indicated in Table 2, the overall sulfur recovery in Case I is 95.46 which is just 0.06% less than the recovery in Case II. Therefore, the effect of producing  $H_2O$  in the inline burners on the overall sulfur recovery can be neglected.

Moreover as mentioned above, burning the fuel gas will increase the amounts of  $CO_2$  in the tail gas stream which has a negative effect on the selective absorption of  $H_2S$  in the absorber column of the TGT section. Although the component molar flow rate of  $CO_2$  is increased in Case I rather than Case II from 268 kmole/h to 274 kmole/h, according to data presented in Table 2 the concentration of  $CO_2$  in the tail gas stream is decreased in Case I rather than Case II which is related to the increasing of  $N_2$  content in the tail gas stream. Therefore,  $CO_2$  concentration is not considered a negative point in Case I rather than in Case II.

Table 2. Comparison of Claus	section in Case I	and Case II	(Without TGT)
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Property	Case I	Case II
Required utility, kg/h	110	2407
Utility cost, thousand \$/year	154	152
First reactor inlet stream Std Flow, m <sup>3</sup> /h	30905	29701
Second reactor inlet stream Std flow, m <sup>3</sup> /h	30750	29107
Tail gas std flow, m <sup>3</sup> /h	30518	28876
Overall sulfur recovery	95.46	95.52
$CO_2$ in tail gas, mole%	21.27	22

In the next step, the TGT section is also taken into consideration and the acid gas from the top of the TGT regenerator column (stripper) is recycled to the entrance of the Claus section (see Figure 1 and Figure 2). The same condition is supposed in the TGT section for both cases. For example, the flow rate of the Amine solution is equal to 210 m<sup>3</sup>/h, as well as the reboiler duty is set to 10.67 MW. A similar analysis is performed for this case and the results are presented in Table 3. As indicated in this table, the required utility cost is approximately the same yet. The amount of increase in the flow rate of the Process gas entering the first reactor, the second reactor, and the output gas from the Claus section, in Case I compared to Case II, is 4.4%, 6%, and 6.1, respectively. These results show that considering inline burners for reheating the process gas will increase the required investment for Claus and TGT sections. Although the Claus section alone has less recovery in Case I compared to Case II, similar recoveries are achieved when the TGT section is applied. Like the previous simulation, in the TGT active mode, the CO<sub>2</sub> concentration of the tail gas stream in Case I is smaller than in Case II.

Property	Case I	Case II
Required utility, kg/h	124	2689
Utility cost, thousand \$/year	174	170
First reactor inlet stream Std flow, m <sup>3</sup> /h	34146	32698
Second reactor inlet stream Std Flow, m <sup>3</sup> /h	33984	32045
Tail gas Std flow, m <sup>3</sup> /h	33729	31791
Overall sulfur recovery	99.93	99.93
CO <sub>2</sub> in tail gas, mole%	25.29	26.11

Table 3. Comparison of SRU (Claus and TGT) in Case I and Case II

The achieved results show that the investigated reheating methods have not any effect on the performance of SRU and similar sulfur recoveries are achieved in both cases. Although applying inline burners has benefits such as lower pressure drop and exact control of the temperature, especially for the first reactor, because of the larger amounts of process gas, using inline burners needs more investment rather than using steam heat exchangers. Moreover, the exact controlling of the air in the inline burners is difficult in operation, and severe corrosion is reported in some refineries using inline burners due to oxygen leakage.

# 3. Conclusions

Reheating of the process gas with inline burners is compared with steam heat exchangers. The achieved results showed that reheating methods do not have any effect on the performance of SRU and the recovery of sulfur. On the other hand, using inline burners need more investment rather than using steam heat exchangers due to burning fuel gas and larger amounts of process gas. Moreover, the exact controlling of air in the inline burners is difficult in operation, and severe corrosion is reported in some refineries using inline burners due to oxygen leakage. As a result, it is advisable to avoid the inline burner method to reheat process gas in the sulfur plant.

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To whom correspondence should be addressed: Hamid Reza Mahdipoor, Gas Transportation and Processing Technology Development Division, Research Institute of Petroleum Industry, Tehran, Iran; <u>e-mail: mahdipoor@gmail.com</u>

# Article

# **Open Access**

Pore Pressure Prediction and Fracture Pressure Estimation using Well Logs and Seismic Data from Cobalt Field, Offshore Niger Delta

Mpara Carine Bonkga <sup>1\*</sup>, Olugbenga A. Ehinola <sup>1,3</sup>, Kennedy F. Fozao <sup>2</sup>, Yinka A. Olayinka <sup>3</sup>, Olutayo Y. Lawal<sup>3</sup>, and Oladotun A. Oluwajana <sup>1,4</sup>

- <sup>1</sup> Pan African University of Life and Earth sciences, Department of Geology, University of Ibadan, Ibadan, Nigeria
- <sup>2</sup> Department of Petroleum Engineering, NAHPI-School of Engineering, University of Bamenda, Cameroon
- <sup>3</sup> Energy and Environmental Research Group, Department of Geology, University of Ibadan, Ibadan, Nigeria
- <sup>4</sup> Department of Earth Sciences, Adekunle Ajasin University, Akungba-Akoko, Nigeria.

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#### Abstract

The work is aimed at predicting shale pore pressures in order to identify the different pressure zones and estimate fracture pressures in some wells in COBALT Field, Niger Delta. To achieve this, geoscientific software was used to analyze the well log data. The data was quality checked and true vertical depth computation was carried out to facilitate overburden stress estimation. The extrapolation method was used to compute the overburden stress, through the merging of the bulk density log and the synthetic bulk density that was generated. Using a gamma ray log, mechanical stratigraphy for lithology delineation was carried out to delineate shales from sands. Normal compaction trendline was applied to sonic and resistivity logs using Eaton's method together with Bower's sonic method to predict pore pressures. The Mathews and Kelly's empirical method was applied to vertical stress and pore pressures obtained to estimate fracture pressures. Also, Petrel software was used for seismic interpretation, after which time and depth maps were generated. The tops of overpressure ells were incorporated into structural maps to show the distribution of the tops of overpressure wells. It was observed that Wells Cobalt- 01 and 06 are normal pressure wells with the highest-pressure values of 8.61 and 8.67 ppg, respectively; while Wells Cobalt-02, 03, 04, and 05 were over-pressured with values of 10.9, 9.5, 10.5, and 10.01 ppg respectively, with corresponding depths of 1288, 12057, 9674, and 1019 FTTVD, respectively. The fracture pressure for each well was also estimated from the pore pressures. The main mechanisms responsible for overpressure in the COBALT field were identified as loading and unloading, with a small contribution from fault-related lateral pressure transfer. The results observed from the depth map show that the top of overpressure wells is distributed or compartmentalized into different fault blocks. The outcome of this work could facilitate proper planning and drilling of future wells in the field.

**Keywords**: Pore pressure; Fracture pressure; Overburden stress; Normal compaction trendline; Eaton's method.

#### 1. Introduction

Pore pressure is the pressure acting on fluids within the pores of a reservoir <sup>[1]</sup>. Pore pressure is also referred to as formation pressure. Technology has facilitated a lot of things and made it easier to achieve various ways of acquiring information on pore pressure before embarking on a journey of petroleum activities such as drilling and production, predicting any sudden expectations and looking for ways to overcome such challenges either at high-or low-pressure zones. Incomplete knowledge and information about Formation pressure while engaging in drilling activities has led to a lot of devastating situations such as loss of lives,

equipment, and uncontrollable flow of oil onto the surface. If high-pressure zones are accurately predicted and detected, many accidents will be avoided and drilling time will be reduced. Predicting the pore pressure of reservoirs in a field is a major component of exploration, exploitation, and development risk analysis <sup>[2-3]</sup>. Knowledge of Formation pressure is very important in the development of oil and gas fields. Petroleum geologists' ability to determine likely traps, seals, and map hydrocarbon pathways by accessing trap design and basin modelling is dependent on their understanding of reservoir pore pressure <sup>[4-8]</sup>.

Also, formation pressure prediction or pore pressure prediction is critical and very necessary because it facilitates decision-making in well planning and operations, safety conditions for personnel in charge, as well as equipment in use. The outcome from pore pressure prediction determines casing design and drilling fluid to be used during drilling. Without information and knowledge on pore pressure, certain problems are likely to occur, such as kicks, influx, and blowouts. On the other hand, if this information is available, pre-preparations and better planning will be done to overcome such challenges.

# **1.1. Previous studies**

Pore pressure prediction using shale parameters acquired from well log data was first investigated by <sup>[9]</sup>. They calculated overpressures by plotting acoustic velocity and resistivity data from shale in Texas and Louisiana, USA. They showed that resistivity had a normal trend with depth while sonic wave travel time gave a logarithmic relationship with depth in the normally pressured interval. According to <sup>[10-12]</sup>, this pressure-depth relationship measured with a sonic and resistivity log is known as the normal compaction trend (NCT). Any deviation in measured parameters from the usual trendline was used in this approach. Many researchers have successfully predicted pore pressure using resistivity, sonic transit time porosity, and other well log data. Pore pressure research is mostly predicated on the concept that any change in normal pressure causes changes in petrophysical parameters like compaction, porosity, and fluid motion. This indicates that observable metrics that can demonstrate these changes can be used to interpret and quantify pore pressure <sup>[13]</sup>. The pore pressure and the fracture gradient define the mud weight that is required during drilling. Whereas <sup>[14]</sup> provided an equation that accounted for overburden stress and hydrostatic pressure in his work to predict pore pressure using a normal compaction trend line and observed acoustic sonic wave travel time. This equation also uses resistivity data, and this takes into consideration a single NCT for the entire depth of the well and assumes one overpressure origin, which is disequilibrium compaction or undercompaction, but most times empirically modified to account for multiple mechanisms.

Bowers presented a method different from Eaton that considers disequilibrium compaction and an unloading effect that results from fluid expansion <sup>[15]</sup>. He showed that a drop in sonic velocity without a decrease in bulk density could come from an unloading effect. He carried out this work in many Formations using data from the Gulf of Mexico and derived effective stress from pore pressure and overburden stress based on sonic log data.

Both Eaton and Bowers <sup>[14]</sup> and <sup>[15]</sup> methods are based on the fact that porosity decreases with burial depth in a normally pressured Formation or hydrostatically pressured Formations. Also, areas with overpressure usually reduce the tendency for porous rocks to compact, leaving them with high porosity. Swarbrick conducted his research and provided some porosity method lobe holes <sup>[16]</sup>.

A lot of challenges are faced in the petroleum industry, such as increased nonproductive time, stock pipe, formation fracture, wellbore instability, kicks, blowouts, loss of lives, and damaged equipment. This is made worse because accurate information is not readily available to petroleum drilling companies. Again, most problems also arise from the drilling fluid in which mud weights are not accurately designed and also from the fact that casing designs do not match the required depth or are not properly selected. Furthermore, most challenges are not well managed due to inadequate risk preparation and poor incident response. Also, inadequate monitoring or attention, which sometimes results from the staff. Lastly, even though unavailability of data is often the problem, most of the time data is not available to do the necessary checks required before embarking on an activity such as drilling.

In the report titled Loss of well control occurrence and size estimation by <sup>[17]</sup>, his report showed that between 2000-2015, 45% of Loss Of Well Control (LOWC) reported in the SINTEF Offshore blowout events occurred during drilling. And Nigeria recorded 2 exploration drilling blowout events. This means that this problem is not just Nigeria's but a worldwide problem. Therefore, this study will therefore provide the pressure distribution in this field "COBALT" and the cause of the pressure, which will aid with information that would be of importance to the drilling and production industry.

The aims and objectives of this study are to predict pore pressure and identify geopressured zones from well logging and seismic data. That is to identify normal, subnormal, abnormal, and overpressure zones. Also, an estimation of pore and fracture pressures were made to determine the influence of pore and fracture pressure on wellbore stability and production of maps for the top of overpressure wells to observe the effect of faults.

# 1.2. Geological setting

The Cobalt field is found in the offshore portion of the Niger Delta Basin (Figure 1). The Niger Delta is a prolific basin that is ranked among the topmost producing basins in the world and Africa at large. It is found at the apex of the Gulf of Guinea on the West African continent <sup>[18]</sup>, which is part of the triple junction formed in the Cretaceous during the continental breakup.



The Niger Delta Basin is bounded in the west by the Dahomey basin, in the east by the Cameroun volcanic line/Abakaliki fold belt, in the north by the Anambra Basin, and in the south by the Gulf of Guinea 4000m bathymetric contour. It is situated in the southern part of Nigeria, between longitudes 3–90E and latitudes 4–60N. It is made up of depobelts that form one of the largest regressive sequences in the world and a highly prolific hydrocarbon province delta with an area of about 300,000 km<sup>2</sup> [18-19]. According to <sup>[20]</sup> Hospers 1965, the Niger Delta is comprised of 500,000 km<sup>3</sup> of sediments, covers an area of 75000 km2 and consists of a 12 km thick sedimentary structure that indicates a progradational package [18-19,21-22]. A lot of studies

Figure 1. Location and distribution of wells in study area

have been carried out by many researchers concerning the evolution and stratigraphy of this basin.

The geodynamics of the separation of the African and South American continents, as well as the tectonics of the development of the Benue Trough during the Late Jurassic, are intimately tied to the evolution of the Niger Delta.

# 2. Materials and methods

The following datasets were used: The dataset for this project was provided by a petroleum company in Nigeria. The datasets provided were well logs for six wells (Cobalt 1 to 6), deviations, checkshots for all six wells, and 3D seismic data in the Cobalt Field. Figure 2 shows the

workflow adopted to achieve the objectives of this study. The methodology adopted to realize the objectives of this work is described in Figure 2 and explained as follows.



Figure. 2. Workflow adopted to achieve the objectives of this study

# 2.1. True vertical depth (TVD) computation

The TVD computation is essential for conversion from measured depth (MD) to TVD since the overburden stress needs to be calculated for the entire depth. This conversion was done for each well. TVD computations are important in the determination of bottom hole pressures, which are caused partly by the hydrostatic head of fluid in the wellbore.

#### **2.2. Overburden stress or lithostatic pressure**

The extrapolation method was used in this work to calculate overburden stress. The density profile was built at depth with the logged bulk density, then the composite or synthetic bulk density was built using the assumed salt density and densities of the formation. Seawater density is considered at 1.03g/cm3, which is essential for the overburden stress calculation. Therefore, by integrating this synthetic density with logged bulk density, overburden stress (equation 1) and overburden gradient were obtained.

 $S = \rho_f gz \dots \dots$ 

(1)

where z,  $\rho_f$  and g are the height of the column, the fluid density, and acceleration due to gravity, respectively.

# 2.3. Mechanical stratigraphy and well correlation

The mechanical stratigraphy flag is done by using gamma Ray for lithology discrimination to delineate between sands and shales. The Gamma Ray index (GRI) was used to calculate the volume of shale (equation (2)). The Gammar Ray measures the natural radioactivity in the formation and it is this property which helps to identify the lithologies.

$$IGR = \frac{GRlog-GRmin}{(GRmax-GRmin)}.$$

(2)

where: IGR = Gamma ray index ;GRlog = Gamma ray estimation from the GR log in the zone of interest ;GRmin = Minimum gamma ray for the clean sand ;GRmax = Maximum gamma ray for the shale.

#### **2.4.** Pore pressure prediction

The Eaton Resistivity and Bowers method were used because the Niger Delta is a Tertiary Basin and the main processes that lead to the generation of overpressure in these Tertiary Basins (Gulf of Mexico, Niger Delta) are rapid deposition, subsidence, and burial in low permeable sediments <sup>[23-25]</sup> such as shales, causing mechanical disequilibrium compaction, which is a function of effective stress.

# 2.4.1. Eaton's sonic velocity method with depth-dependent normal compaction trendline

Pore pressure gradient prediction from sonic compressional transit time ( $\Delta tn$ ) was achieved based on [14] Eaton's (1975) empirical equation (equation 3).

$$Ppg = OBG - (OBG - Ph) \left(\frac{\Delta tn}{\Delta t}\right)^m \dots$$

where  $\Delta t$  is the transit time in shales obtained from the well log;  $\Delta tn$  is the transit time in shales at the normal pressure; Ph is hydrostatic pressure; OBG is the overburden stress; *m* is an exponent, and normally the exponent; *m* = 3 can be applied in case the overpressure was generated by undercompaction without any secondary mechanism of pore pressure generation.

The transit time  $(\Delta tn)$  is determined from the normal compaction trendline in normal pressure conditions, and it is obtained by applying in Eaton's sonic method.

#### 2.4.2. Eaton's resistivity method with depth dependent normal compaction trendline

The same equation (Eaton's equation) was used, and the same input parameters were applied except that sonic wave travel time was replaced by resistivity log as shown in equation (4)

(4)

$$PPg = OBG - (OBG - PPN) \left(\frac{R}{R_N}\right)^{\lambda} \dots$$

where `PPg' is the pore pressure gradient (ppg); `OBG' is the Overburden gradient (ppg); `PPN' is regarded as the normal pore pressure gradient (ppg); "R" is the observed resistivity (ohms-m); "R<sub>N</sub>" is the Normal Resistivity (ohms-m); and "x" is the Eaton exponent, which is 1.2 and the Eaton Resistivity exponent of 1.

In Eaton's equation, there is always a problem in determining the shale resistivity at hydrostatic pore pressure. This calls for the need for a compaction trendline for pore pressure prediction. Rn, which is the normal resistivity function of burial depth, can be calculated from the normal compaction trend line using equation 5.  $Rn = R_0 e^{bz}$ (5)

By substituting the equation (5) into equation (4), Eaton resistivity equations can be written:  $Ppg = 0BG - (0BG - Png) \left(\frac{R}{R_0 e^{bz}}\right)^n \dots$  (6)

where 'R' is the shale resistivity measured at depth 'Z'; Ro is the normal compaction shale resistivity in the mudline and 'b' is the logarithmic resistivity normal compaction line slope; Png is the normal or hydrostatic pressure; OBG is overburden gradient.

# 2.5. Bower's original method

Compressional velocity was determined in this work using Bower's equation (equation.7)  $Vp = V0 + A\sigma e^{B} \dots \dots \dots$  (7)

where vp is the compressional velocity at a given depth; V0 is the compressional velocity in the mudline (i.e., the seafloor or the ground surface, normally V0l is approximately 5000 ft/s, or 1520 m/s) so 5000m/s was used; A and B are the parameters calibrated with offset velocity versus effective stress data,  $\sigma e$  is effective stress.

#### **2.6.** Fracture pressure determination

In this work, Matthews and Kelly's <sup>[26]</sup> method was used for the estimation of fracture pressure (Figure.3.4.). That is fracture pressure (equation 8) gradient is given as

#### $FG = k_1 (S - P) + P \dots \dots$

(8)

where S is the overburden stress gradient; P is the pore pressure gradient, and  $k_1$  is the matrix stress or effective stress coefficient.

#### 2.7. Well correlation and seismic section interpretation

The correlated reservoirs were used as the top of sand on the seismic section, which corresponded to a particular amplitude to be mapped. The 3D seismic data of the Cobalt field was provided in SEY format and was loaded onto Petrel 2017 software. Before loading the seismic data on petrel, the project setting was done to carry out coordinate referencing and to set the data to correct units. After loading the data, it was confirmed to be fine through a quality check. The loaded seismic was realized to reduce the volume for easy interpretation or to see the structures well.

The structural interpretation, which consists of fault mapping to get a good understanding of the structural framework of the Cobalt field, was carried out. Structural smoothening was used to enhance the seismic reflections for better interpretations of the faults. The variance attributes (displayed on a time slice) helped in the understanding of the fault trend. This attribute helps increase the clarity of the seismic data for easy and accurate interpretation.

The seismic to well tie was performed using a corrected sonic log and well check shot data. The Checkshot is used to calibrate the relationship between well depths and times calculated from a sonic log. Well tie allows well data, which is in depth, to be compared to seismic data, which is in time. This helps to generate a synthetic seismogram using a Ricker wavelet, which has a good fit and further helps to pick the horizon on the seismic section. The horizons were mapped across the inline and crossline to produce time and depth maps.

The horizons were neatly picked across inline and cross lines, after which they aided in the production of fault polygons. Four horizons were picked, corresponding to the top and base of each reservoir. From the fault polygons, boundaries were generated, which led to the production of time-structural maps.

Time structural maps were generated for the four surfaces from the integration of the fault polygons, boundary polygons, and each horizon. After that, time structure maps were converted using the function to generate depth structural maps to show the distribution of top of overpressures.

#### 3. Results and discussion

From the analysis gotten from well log and seismic data, the overburden stress, the mechanical stratigraphy flag, pore pressure prediction, and fracture pressure together with seismic interpretation are explained in detail.

#### **3.1. Overburden stress**

The overburden stress is increasing with an increase in depth. The overburden stress and depth readings all begin with 0 ft, 0 psi, and the overburden stress ranges from a minimum of 0 psi to 111130.1 psi as the maximum, which is in Well one. The table (Table 1) shows the minimum and maximum overburden stress for each well and the minimum depths at which the overburden stress value begins to change from zero psi (0 psi). Overburden stress was found to increase with increasing depths into the subsurface.

Well Name	Depth(ft)	Overburden stress min (psi)	Depth(ft)	Overburden stress max (psi)
Cobalt_01	69.5	0.22	11117.5	11130.1
Cobalt_02	74.5	0.22	12336	1089.02
Cobalt_03	72.5	0.22	12120	11119.50
Cobalt_04	95.5	0.22	10300	8815.95
Cobalt_05	116	0.22	10160	9126.63
Cobalt_06	108	0.22	1167	8898.45

Table 1 Minimum and maximum values o overburden stress

Well name	Top of over- pressure(ft)	Start pres- sure(ppg)	Max depth(ft)	Max pressure. (ppg)	State
Cobalt_01		7.73	11117.5	8.61	Normal pressured
Cobalt_02	11140	8.61	1288	10.9	Over pressured
Cobalt_03	11437	8.61	12057	9.5	Over pressured
Cobalt_04	9663.5	8.57	9674	10.5	Over pressured
Cobalt_05	9555	8.55	1019	10.01	Over pressured
Cobalt_06		7.73	1067	8.57	Normal pressured

Table 2. Results of pore pressures shows top of overpressure wells for overpressured wells and maximum pressures of each well

	Table.3.	Minimum	and	maximum	fracture	pressures
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Well Name	Depths(ft)	Minimum fracture pressure(ppg)	Max(ft)	Maximum fracture pressure(ppg)
Cobalt-01	3119	13.27	1081	16.61
Cobalt-02	4585.5	12.27	12307	16.61
Cobalt-03	3441	11.3	12057	15.6
Cobalt-04	4774	12.86	10025	15.01
Cobalt-05	4887	13.50	10144	15.43
Cobalt-06	7606.5	14.08	10778.5	15.06

# 3.2. Mechanical stratigraphy

The mechanical stratigraphy shale flag is shown below (Figure.3.1a-f) for all the wells, respectively, showing the lithologies for each well. Their results showed that thicker sequences of sand are at shallower depths up to 5000 ft and above are intercalations of sands and shales, up to 9000 ft and above 9000 ft are made of thicker sequences of shales, thin clays, and small pockets of sand. This reflects the stratigraphy of the Niger Delta, where the youngest is the Benin Formation consisting of continental sands and gravel, the Agbada Formation, which is a paralic sequence of interbedded sandstones and shales, and lastly, the Akata Formation, which is predominantly shales and clays with some turbidites and small sand bodies.



Figure. 3.a-f. Display of lithologies for the six wells respectively

#### 3.3. Pore and fracture pressure prediction

In Cobalt\_01 and Cobalt\_06, pressure values begin at 7.73ppg and increase with depth, with the normal pressure of 8.61ppg and an Eaton resistivity value of 8.61ppg for Cobalt\_01. These pressure values are within the normal pressure of 0.433 psi/ft or 8.5 ppg and wells with predicted pore pressure values are considered normal pressure wells or hydraulically pressured wells. Eaton's Resistivity obtained values for shallow depths were as high as 8.7 ppg but were not considered because the shallow depths contain fresh water and fresh water has high resistivity. The resistivity log is not good to be used alone since the resistivity log is affected by many factors such as salinity, anisotropy, borehole diameter, drilling fluid, lithology, and temperature. Therefore, corrections are needed when these effects are profound. Also, according to <sup>[27]</sup>, pore pressure near the wellbore is affected by the induced stresses and, therefore, deep resistivity is used for calculating pore pressure. The pore pressure within this range of 8.61 ppg shows that this well is a normal pressure (Table 2) as this is illustrated in the line plot (Figure. 4a) and the normal trend is shown in the cross plot (Figure. 4b).



Figure 4 a-b Line plots displaying overburden pressure with Eaton's resistivity pore pressure, normal pressures and fracture pressures with depth showing normal pressure for Cobalt\_01 and Cobalt\_06 respectively

The fracture pressure ranges from 13.27 ppg at 3119 ft. and 16.61 ppg at 10981 ft. and it is also shown in Table 3. The fracture pressure is normally considered as the upper or maximum bound of the drilling mud weight. If the drilling mud weight exceeds the formation, it will crack and the drilling fluid will get lost into the formation. Information about fracture pressure also important to select the casing design for wells.

Cobalt\_06 has the same characteristics as Cobalt\_01. The pressure values start at 7.73 ppg and increase hydrostatically up to 11067 ft with a pressure of 8.57 ppg for Eaton resistivity obtained value (Figure. 4b). This pressure is within the normal pressure range which is about 8.5 ppg or 0.433 psi/ft. There is a Sonic log present for this well, but it is not good enough to be used and the sonic log is incomplete as it is difficult to draw a normal compaction trendline.

Pore pressure values including those of Cobalt-06 are presented on Table 2 while those of fracture pressure are presented on Table 3. The fracture pressure values range from 14.08 ppg at 7666.5ft to 15.06 ppg at 10778.5ft. As mentioned earlier, the fracture pressure, which is considered the upper or maximum bound of drilling mud weight. these values will help to design the maximum mud weight and casing setting here. Line plots are displayed (Figures 4a and 4b) to show pressure plots for normally pressured or hydrostatic wells.

Wells with high pressures are shown as the Eaton Resistivity and Eaton Sonic methods together with the Bowers method, are tracking each other and have higher values that are greater than hydrostatic pressure (Cobalt\_02, Cobalt\_03, and Cobalt\_04). While for wells that have only Resistivity log (Eaton's resistivity method), the pore pressure values are high for overpressure wells as seen in Cobalt\_05.

In Cobalt\_02, the pressure values increase hydrostatically with depth. It starts with normal pressures of 8.52 ppg and pressure values start to increase at the depth of 11140 ft and pressure values of 8.60 ppg (2025.641 psi) (Figure 5a) as the top of overpressure. A maximum pressure value of 10 ppg is reached as shown in Figure (5b) at a depth of 11288.5 ft with pressure values of 9.61 ppg (5633.9psi) for Bowers' original, 10.90 ppg (6423.484psi) for Eaton's Resistivity, and 10.17 ppg (5963.343psi) for Eaton's Sonic, also shown in Table 2.



Figure 5a shows predicted pore and facture pressure profiles while 5a line plot shows pressure variations with depths and indication top of overpressure and drilling margin

These pressure values are all greater than 8.60 ppg for normal pressure values. Overpressure is observed as the Eaton's sonic and resistivity methods, as well as the Bowers method, track each other. This is seen as a deviation from the normal pressure. The well appears to have been compacting normally up to a depth of 11140 ft and later had some disturbance which led to a deviation from the normal compaction trend. There is a small reduction in bulk density down to 2.15g/cm<sup>3</sup>, a reduction of resistivity to 1.370hms, and a small increment in the sonic log as well, with intercalation of shales and sands. An increase in pore pressure at this depth could be due to fluid expansion as a result of hydrocarbon generation, while further down, as the pore pressure values increase, it is due to an increase in bulk density of up to 2.6g/cm<sup>3</sup> resulting from undercompaction or compaction disequilibrium where sediments compact and the surfaces seal off such that pore fluid cannot be expelled or is expelled at a slower rate. This situation leads to higher pressure values than normal. Figure 5c is a cross plot of derived acoustic velocity (Vp) against Density log which shows the trend representing the different mechanisms causing the overpressure [<sup>28</sup>]. Cross plot has also been used to show the mechanisms responsible for overpressure in Niger Delta. In addition, the fracture pressure,

which is considered the upper or maximum bound of drilling mud weight for this well. The fracture pressure for this well ranges from 12.27 ppg to 16.61 ppg at 45855 ft to 1230 ft respectively, as presented in Table 3.



Figure 6a shows predicted pore and facture pressure profiles while 6b line plot shows pressure variations with depths and indication top of overpressure and drilling margin



Figure 5c and 6c. Cross plots showing the trends of different overpressure mechanisms for Well 02 and Well 03 respectively.

Also, Cobalt\_03 has similar characteristics as Cobalt\_02 and has the same mechanism causing overpressure. In Wells 03, the pressure begins with a normal hydrostatic pressure of 8.4 ppg and the pressure starts increasing at a depth of 11437 ft. This is seen by Eaton Resistivity pressure value of (8.6ppg or 5114.227psi) and Eaton Sonic pressure value (8.6ppg or 5114.227psi) tracking each other together with the Bowers sonic (8.6ppg or 5114.227psi)

method (Figure 6a). The pressure values increase to 9.0 ppg (5618.858 psi), 10.5 ppg (6567.645 psi), and 9.53 ppg (5937.675 psi) at 12025 ft, respectively, and the maximum pressure value is shown in Table 2. This overpressure is indicated on the line plot (Figure 6b) as top of overpressure (11437 ft), which is the depth at which the pressure starts to increase above normal. These pressure values are greater than the values for a normal pressure well. This well is therefore considered an overpressure well as the pressure at this depth deviates from the normal pressure values of 8.5 ppg. Figure 6c shows a cross plot of derived acoustic velocity (vp) against density log, indicating the trends causing the overpressure in Well 03. The fracture pressure for this well ranges from11.3ppg to15.6ppg at 3441ft to 1205ft respectively as shown on Table 3, and is considered as the upper or maximum bound of drilling mud weight for the well.

In Cobalt\_04, the pressure also begins with a normal pressure value of 8.47 ppg and an increased pressure as from 9663.5 ft with pressure values of 8.66 ppg (4084.401 psi) for Bowers' original, 8.7 ppg (4109.501) for Eaton's Resistivity, and 9.23(4351.695psi) for Eaton's Sonic, which shows a deviation from the normal hydrostatic pressure (Figure 7a). This is also referred to as the top of overpressure (Figure 7a-b).



Figure 7a Shows predicted pore and facture pressure profiles while 7b is line plot of pressure variations with depths and indication top of overpressure and drilling margin





The pressure values reach a maximum (Figure 7b) at 9674 ft with pressure values of 8.89 ppg (4201.614psi), 9.94 ppg (4695.166psi) and 9.4 ppg (4470.188psi) respectively for Bowers original, Eaton's Resistivity, and Eaton's Sonic, the highest pressure value is presented in Table 2. The predicted pore pressure for this well is greater than normal pressure values of 8.5 ppg, therefore well 04 is considered overpressured. This increase in pressure is because of the increased density of sediment, which is caused by undercompaction or compaction disequilibrium where there was rapid sedimentation in such a way that the fluid could not be expelled at the faster rate, which eventually led to overpressure. A cross plot of derived acoustic velocity(Vp) against density log (Figure 7c) is used to represent the trend causing overpressure in Cobalt 04.

The fracture pressure for Cobalt\_04 ranges from 12.86 ppg at 4774 ft up to 15.43 ppg at a depth of 10025 ft, respectively. The fracture pressure, which is considered the upper or maximum bound of drilling mud weight for this well, shown in Table 3. igure 7a Shows predicted pore and facture pressure profiles while 7b is line plot of pressure variations with depths and indication top of overpressure and drilling margin

In addition, in Cobalt\_05, the pressure begins with a pressure that is hydrostatic; that is, pressures of 8.44 ppg, and the pressure increases normally with depth up to 9555 ft with the pressure of 8.61(414.24 psi) for Eaton Resistivity and normal pressure (Figure 8a). It then increases to 10.2 ppg (5075.749 psi) at 10019 ft. The predicted pressure is more than the normal pressure of 8.5 ppg (Figure 8b). This implies that this well is overpressured even though the Sonic log is absent for this well. It would have facilitated the confirmation the pressure values because, as earlier mentioned, the Resistivity log on its own is usually affected by many factors such as hole diameter, fluids, and salinity. It also reads high values for shallow depth as it contains fresh water. This high resistivity at shallow depth is not considered. A cross plot of density log against resistivity log (Figure 8b) shows two trends for the overpressure mechanism, which means that there are two mechanisms that cause overpressure in well 05 (similar Cobalt\_02 and Cobalt\_03). These are undercompaction and fluid expansion. The fracture pressure for Well 05 ranges from 13.50 ppg at 4887 ft up to 15.43 ppg at a depth of 10144 ft, respectively. The fracture pressure, which is considered as the upper or maximum bound of drilling mud weight for this well, is shown in Table 3.



Figure. 8a. Predicted pore pressure profiles, and 8b. Line plot shows pressure variations with depths and indication top of overpressure and drilling margin



Figure 8c. Cross plots showing the trends of different overpressure mechanisms

PPRS\_NORM: Normal predicted pore pressures PPRS EATON S: Predicted pore pressure using Eaton Sonic Method PPRS\_EATON\_R: Predicted pore pressure using Eaton Resistivity Method FPRS\_MK: Predicted fracture pressure using Matthews and Kelly method SVERTICAL: Overburden stress PPRS BOWERS: Predicted pore pressure using Bowers method 1<sup>st</sup>: Primary 2<sup>nd</sup> : Secondary PPP - Predicted pore pressures profiles OBG- Overburden gradients NCT: Normal compaction trendline FP- Fracture pressure TOP- Top of overpressure DM- Drilling Margin

# 3.4. Well correlation

Four identified (hydrocarbon bearing) reservoirs were correlated across from the reading of Gamma ray and Resistivity logs. This correlation helps to better access the continuity and lateral extent of the reservoir (Figure 9).



Figure 9. Well correlation section for the wells

# 3.5. Seismic interpretation

# 3.5.1. Structural interpretation

Fault mapping was the only structural interpretation carried out in the Cobalt Field. The main structures in the cobalt area are the growth fault and rollover anticline, which could be synthetic or antithetic faults that are listric in nature as the fault flattens with depth, as this is typical of the Niger Delta. The major and minor faults were mapped. This was observed by the abrupt discontinuities in reflection horizons and by the sudden change in the vertical displacement of the reflection horizons. One major fault together with 32 minor faults were picked. It is also observed that the hydrocarbon accumulation is around the rollover anticline, which favors the accumulation [18], as most wells are found between them (Figure 10).

# 3.5.2. Seismic to well tie

Seismic to well tie was achieved with the help of checkshot data for each well. Here, the wells, which were in depth, were tied to the seismic in time such that the well tops were superimposed on the seismic section (Figure 11). Each reservoir top corresponds to the peaks and troughs on the seismic section as observed from the main reflectors. These tops were mapped as horizons.

# 3.5.3. Horizon mapping

The horizons were mapped across inline and cross-line and the faults were respected (Figure 10) as it further facilitated the production of subsurface maps.

# 3.5.4. Time and depths maps, Distribution of top of overpressures

Time maps were generated from the mapped horizons, polygons, and boundaries. These time maps were converted using the plot on (Figure 12) to generate depth maps for each sand unit (Figures 12a, 12b, and 12c) as the wells are penetrating the surfaces, while Figure 13 shows the distribution of all the wells on the combined horizon maps. A majority of the wells are seen to penetrate different fault blocks. The mechanism causing overpressure could be as

a result of lateral transfer from one well to another through the faults. The tops of overpressure wells were found penetrating the different faults (Figure 12d). This shows that the top of overpressured wells is compartmentalized into the different fault blocks (12a-c and Figure 12d).



Figure 10. Seismic section showing the mapped faults and sand tops



Figure 11. Calibration plate showing synthetic seismogram







Figure. 12a. Distribution of wells on the depth map for sand top 1  $% \left( 1-\frac{1}{2}\right) =0$ 



Figure. 12b. Distribution of wells on the depth map for sand top 2



Figure 12c. Distribution of wells on the depth map for sand top 3



Figure 12d. Distribution of Top of overpressure wells on the top of overpressure map.



Figure 13 Distribution of all the wells on the combine depth maps

# 3.6. Implication of predicted pore and fracture pressure on wellbore stability

Pore pressure prediction using Eaton's depth-dependent resistivity and sonic with an applied compaction trendline and Bower's method gave excellent pore pressure prediction. This is because the pore pressures reported in other fields in Niger Delta have proven that overpressure mechanism is a result of disequilibrium compaction(loading) and fluid expansion(unloading) especially being Tertiary Basin <sup>[29-33]</sup>. From the results obtained for pore pressure prediction and the estimated fracture pressures for each, each well has a minimum and upper bound mud weight that is required to penetrate the formation. The pore pressures predicted for wells 01, 02, 03, 04, 05, and 06 are listed below. The predicted pore pressure gives the drillers the minimum mud weight required to drill the formation. That is, the mud weight should be designed not to be below the predicted pore pressure. If the well is drilled below this pressure, it could lead to well influx, kicks, which if not handled well, would lead to blowouts, and if the well is drilled above the predicted pressure, it could cause sticking of pipes. The predicted fracture pressure is the upper limit or bound for which if the drilling fluid goes above it, it will fracture the formation, and lost circulation will occur. It can also cause the formation to collapse. That is, the fracture pressure also aids in the selection of the casing that would be required to be placed in the well, because the fracture pressure selects the correct casing to fit a specific depth. This prevents the formation from collapsing, damage (cracks), and gives the borehole stability. Figure 6 depicts the workflow for each well as well as the drilling margin.

# 4. Conclusion

The pore pressure predicted for the six wells in the Cobalt fields shows that four wells were overpressured (Cobalt-02, 03, 04, and 05) while two were normal pressure wells (Cobalt-01 and 06). This means that wells Cobalt-01 and Cobalt-06 will require the drilling mud (fluid) weight to remain between the hydrostatic pressure and not go below or higher than the hydrostatic pressure. For the overpressure wells, Cobalt-02, Cobalt-03, Cobalt-04, and Cobalt-05 the drilling fluid weight should be added to 10.9ppg, 10.5ppg, 9.94ppg, and 10ppg according to the wells respectively to meet these pressure values needed to balance the formation pressure. Their fracture pressures would also help to select the correct casing that will hold the formation to prevent it from instability or lost circulations. Structural interpretation with seismic volume has been able to show the distribution of top of abnormal pressure in the

overpressured wells, hence it can be concluded that these tops of abnormal pressure is compartmentalized within different fault blocks. It can be concluded that, pore and fracture pressures were predicted for COBALT field in Niger Delta Nigeria using well log and seismic data. This information can facilitate proper planning and drilling of future wells in the field.

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To whom correspondence should be addressed: Mpara Carine Bongka, Pan African University of Life and Earth sciences, Department of Geology, University of Ibadan, Ibadan, Nigeria; <u>e-mail: mpara.carine@gmail.com</u>